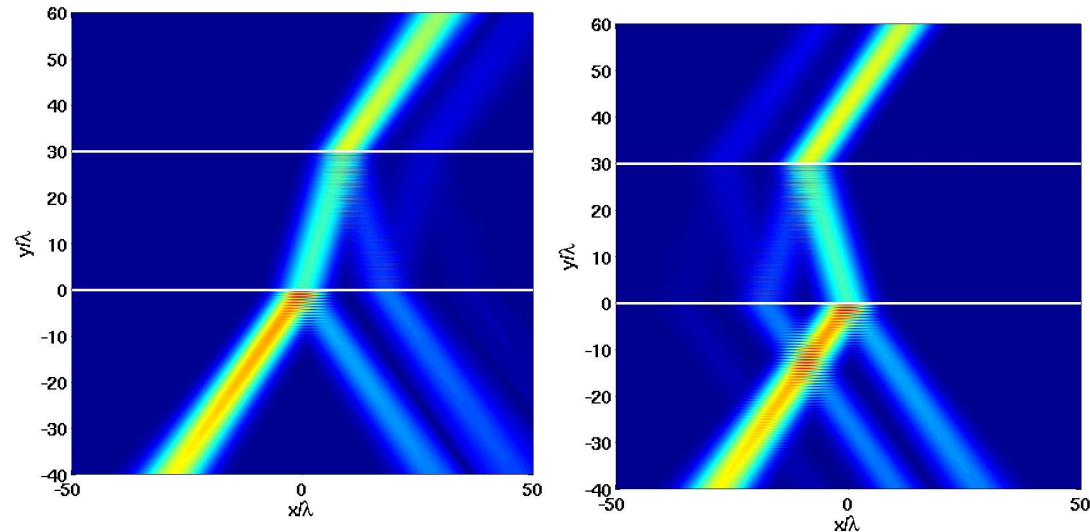




Metamaterials with quantum emitters:

E. Rousseau
G. Cassabois
D. Felbacq

*Laboratoire Charles Coulomb
CNRS- Université de Montpellier
FRANCE*



The team

Didier felbacq

Theoretical studies on:

- electromagnetisms:
- in optics:
- Metamaterials
(homogeneization theory)

G. Cassabois & E. Rousseau

Experimental studies on:

- **In the Near-IR**
- Plasmonics
- Optical Spectroscopy



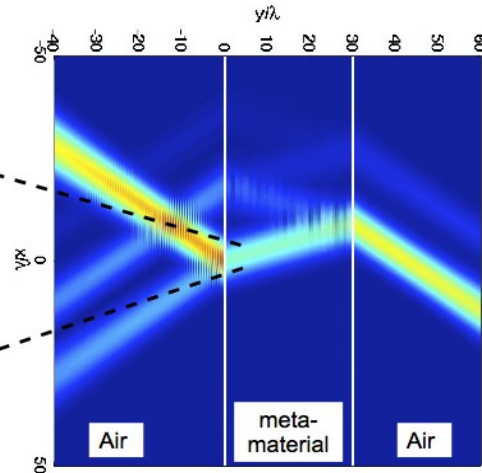
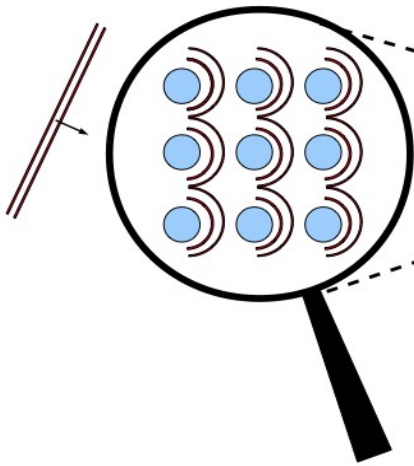
G. Cassabois

D. Felbacq



E. Rousseau

Metamaterials = effective properties



Heterogeneous materials :

@ *microscopic scale*

$$d \ll \lambda$$

Appear like an homogeneous medium

@ *macroscopic scale*

with *effective* optical properties

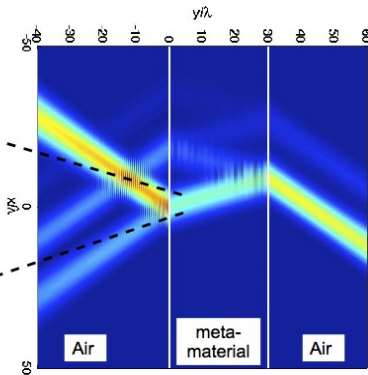
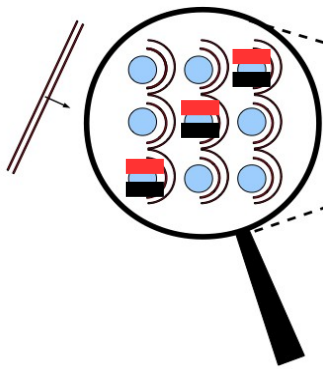
That result from collective effects

@ *microscopic scale*

Not found in Nature:

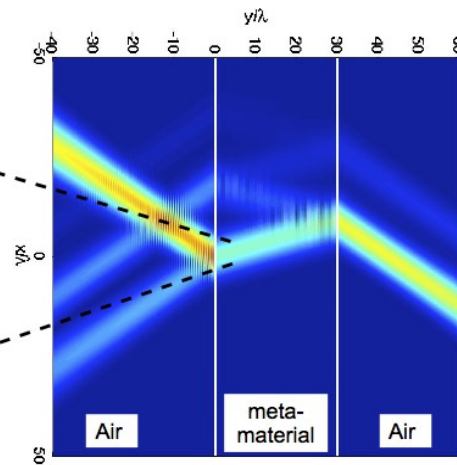
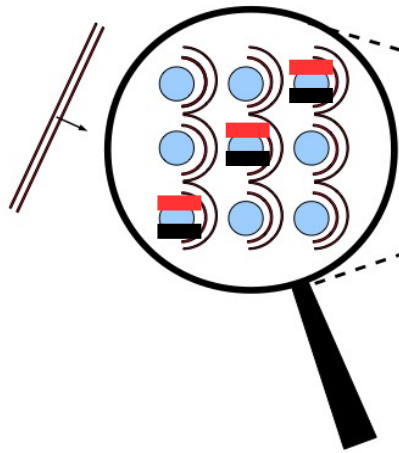
Negative permeability @ the optical frequencies

=> Good but not required.



- Quantum emitters : described by quantum mechanics laws => *already the case?*
- can be prepared in a well known quantum state (at least: initial quantum states)
- correlations exist between quantum emitters (*e.g. The energy level depends on the quantum state of the neighbouring quantum emitters*)
- how light propagation is affected by the emitters quantum states ?

Metamaterials with quantum emitters

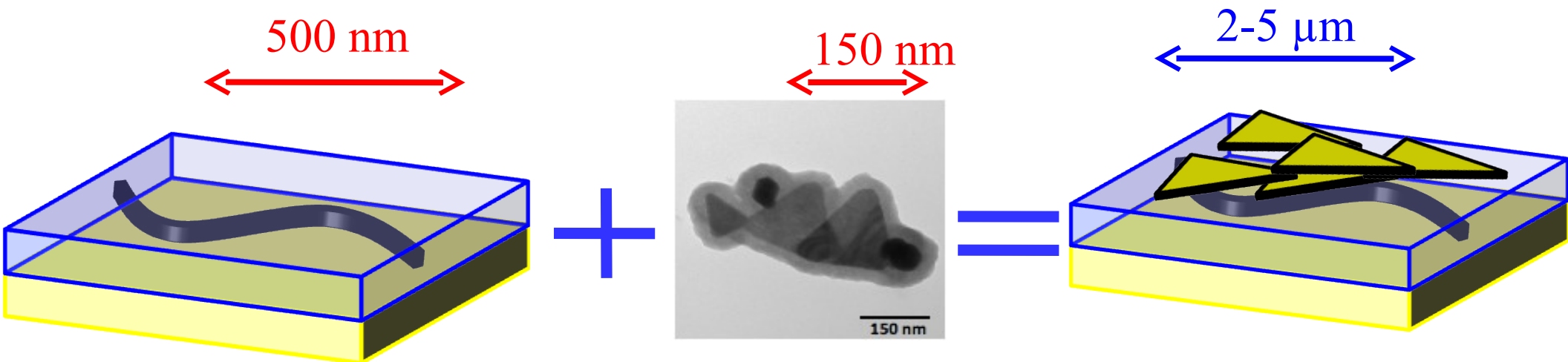


Quantum emitters in a metamaterials structure

Much simpler as a first step

- How the metamaterials affects the behavior of the quantum emitters ?
- Does the quantum emitter affect light propagation ?

Part I : Plasmonic metasurface ?



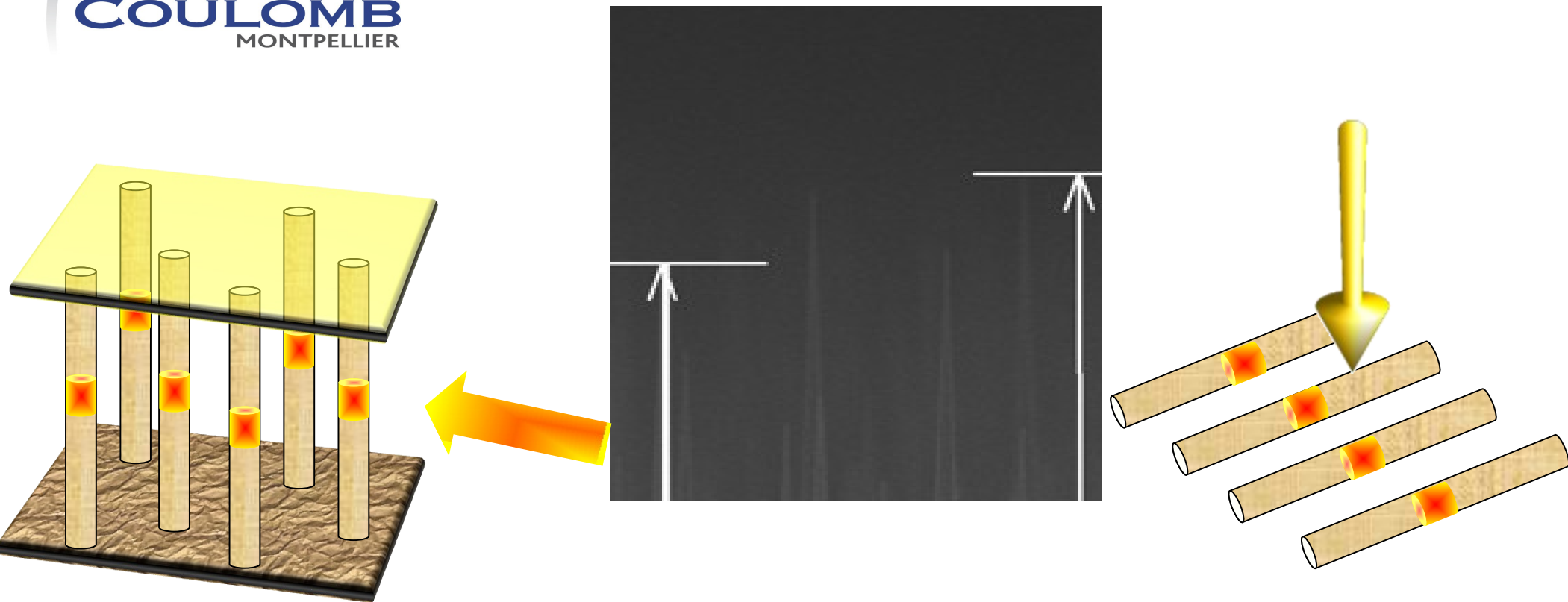
Few carbon nanotubes
(1 single)
Quantum emitter

Collection of
Metallic scatterers

Plasmonic Metasurface ?

Microscopic scale

Macroscopic scale

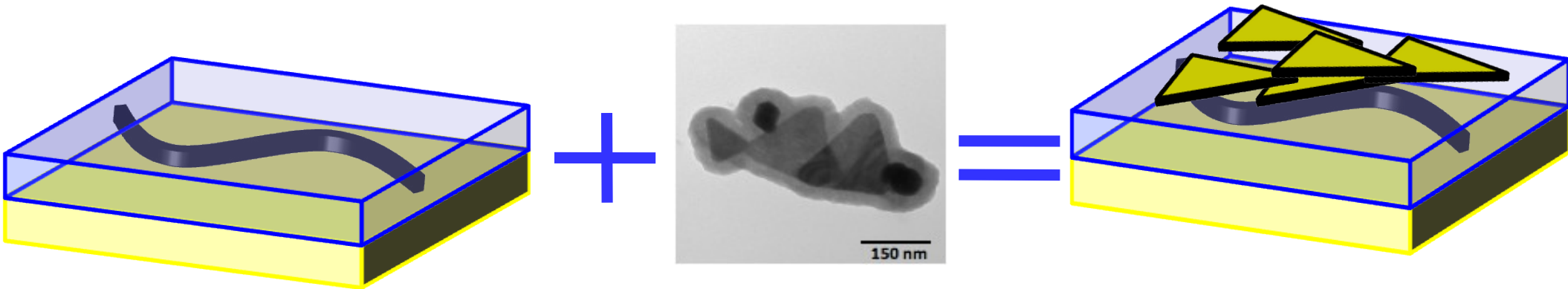


Collection of dielectric rods (InP)

Quantum dots inside each rods (InAsP inclusions)

Metamaterial, metasurface

Part I : Plasmonic metasurface ?



Few carbon nanotubes
(1 single)
Quantum emitter

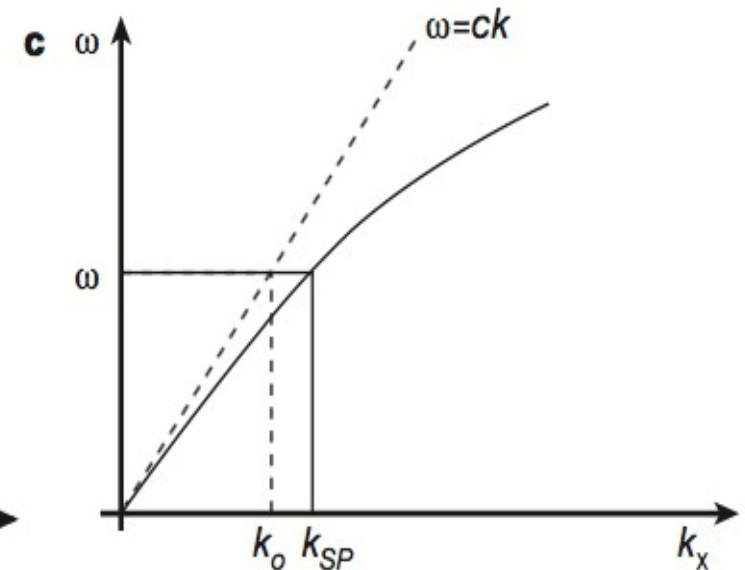
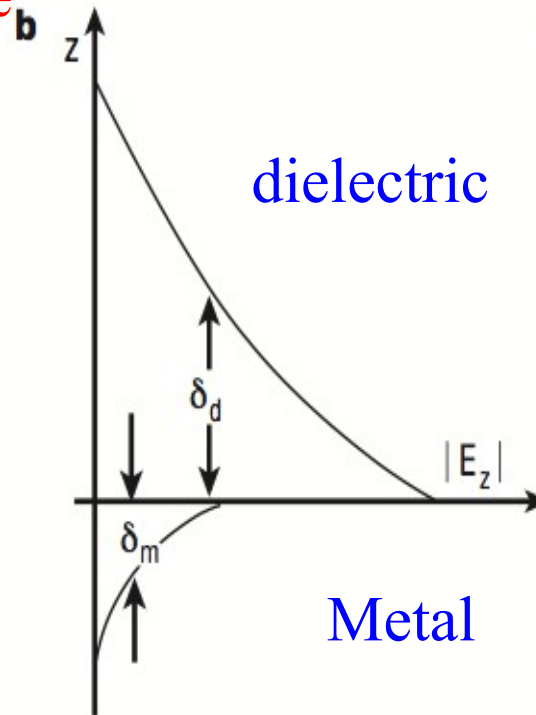
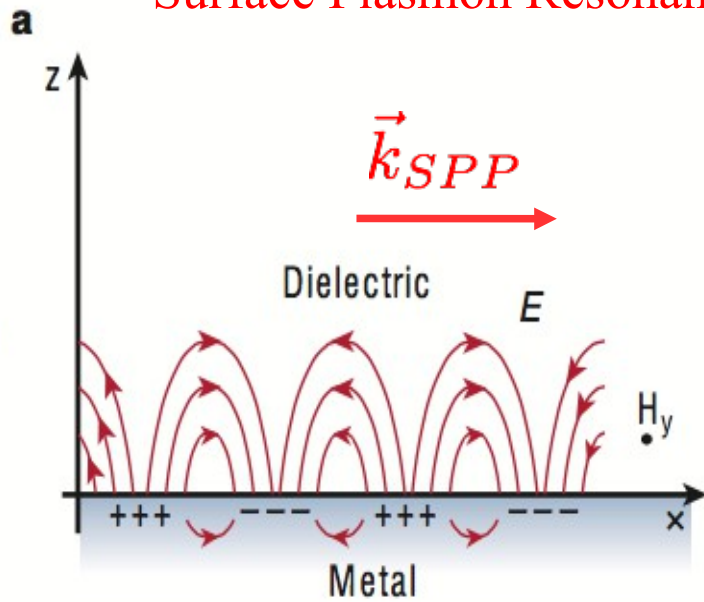
Collection of
Metallic scatterers

Metasurface ?

Field of plasmonics (optics of metals)

Purcell effect : how environment modify quantum emitter luminescence

Surface Plasmon Resonance



Charge density :

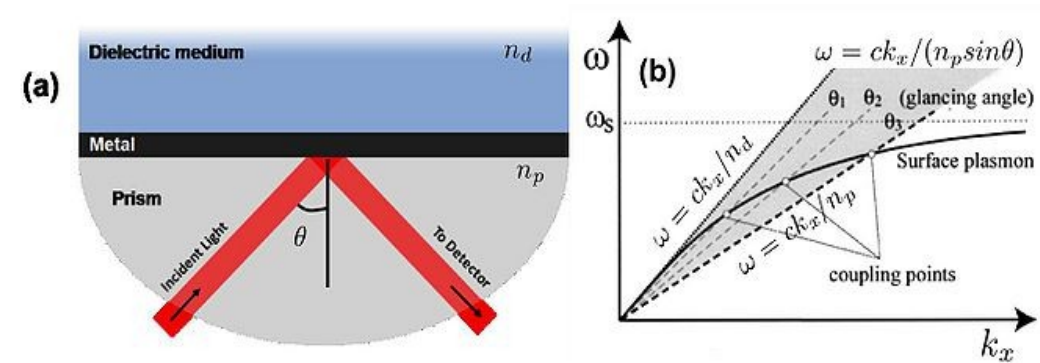
E perpendicular component
Surface wave:
Propagation along the metallic
plane

Evanescent field :

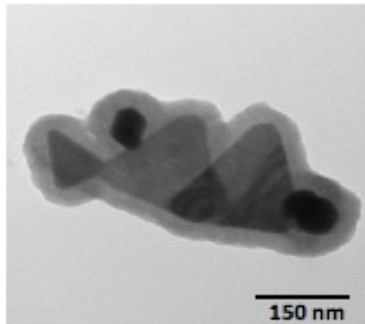
Exponential decrease
from the metallic interface
Field confinement

Dispersion relation :

Below light cone
Non-radiating mode



Prism coupling



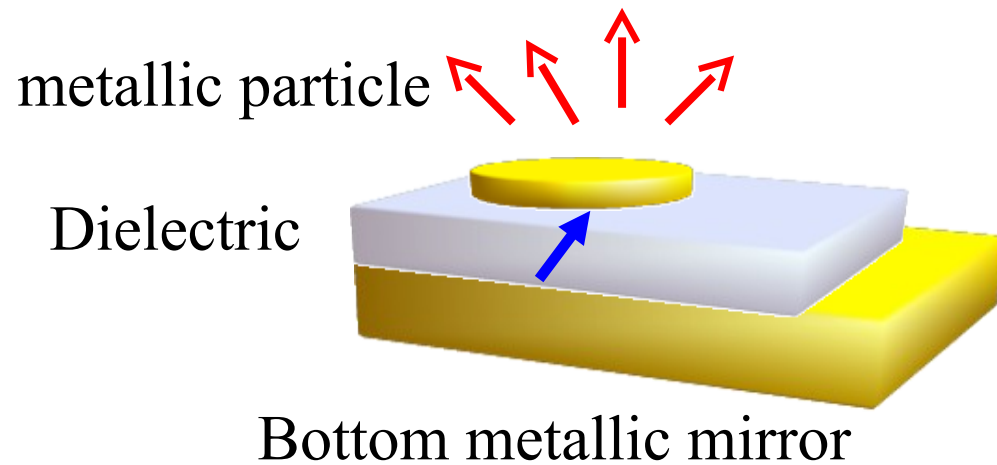
Metallic nanoparticles

Diffraction \Rightarrow Localized plasmon resonance

Can act as antennas in the visible regime :

- modify radiative pattern (directivity)
- modify luminescence intensity (energy extraction, Purcell effect)

The patch antenna



Finite size metallic disk:

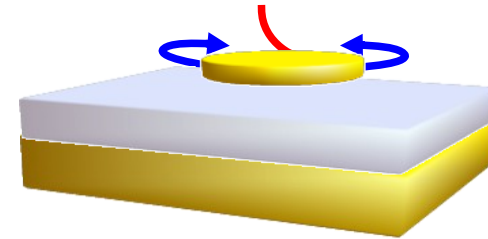
- Can increase of the power radiates by the emitter

Bottom metallic mirror:

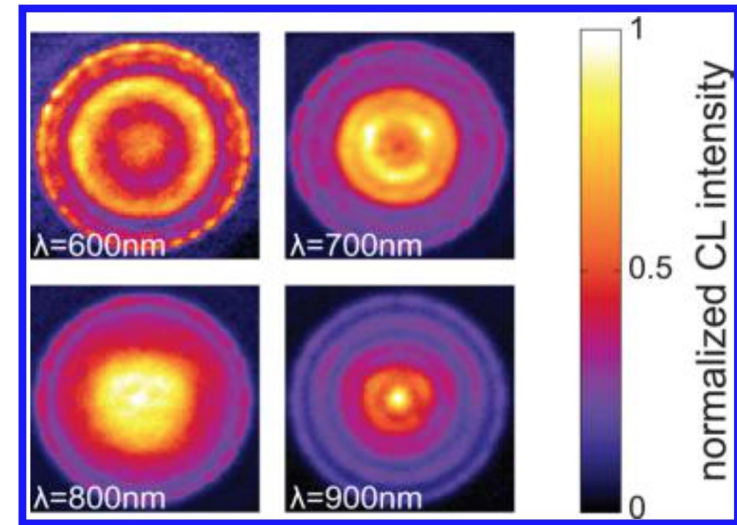
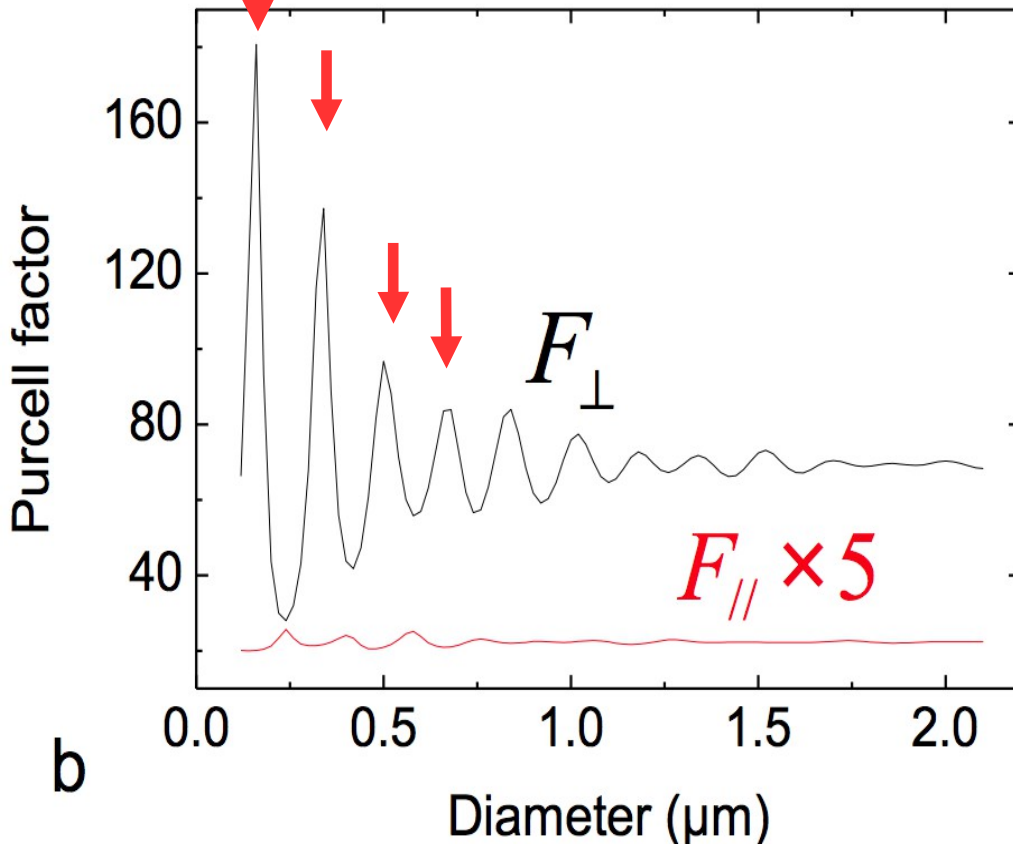
- Light emits in one direction
- Increase electric field confinement



The patch antenna: plasmonic cavity modes



Resonances due to cavity modes

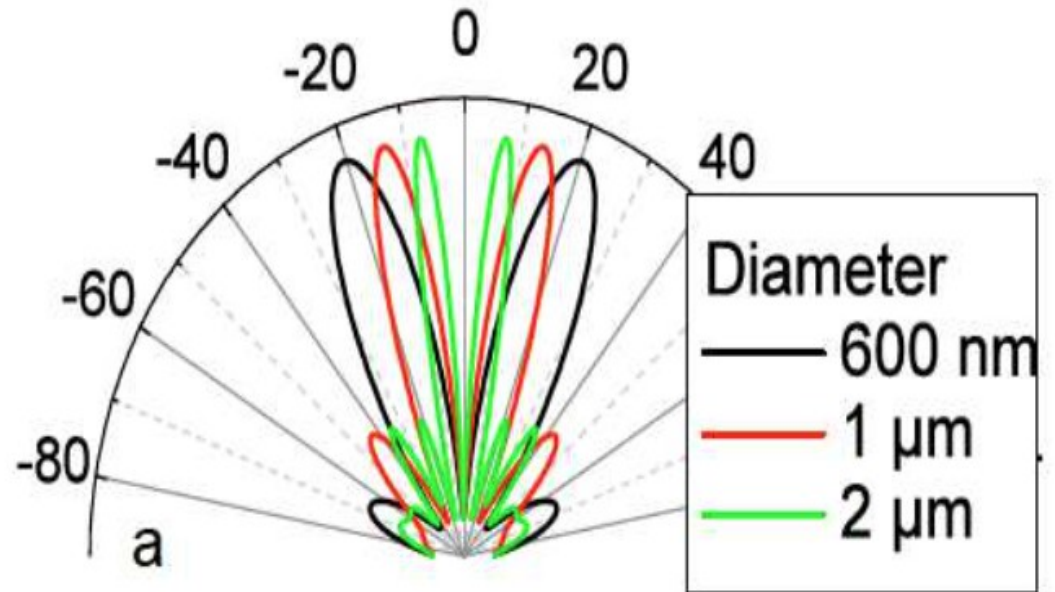
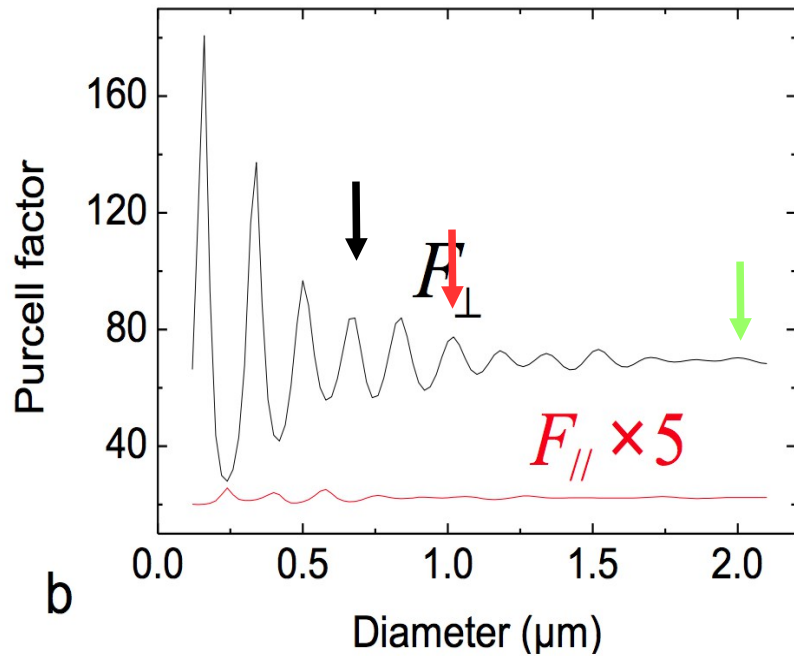
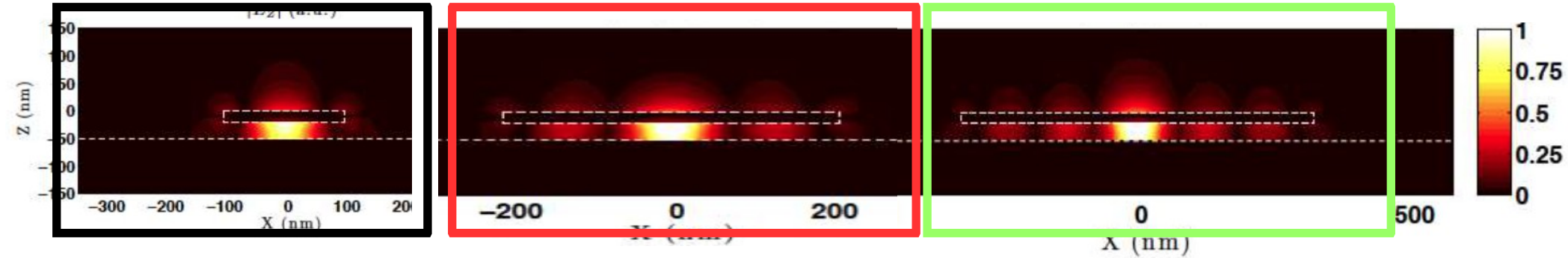


Mohtashami *et al.* ACS Photonics (2014) 1, 1134–1143

Cavity (plasmonic) modes : cavity geometry defines resonant modes
Large cavity (~1μm) many cavity modes (kind of continuum)

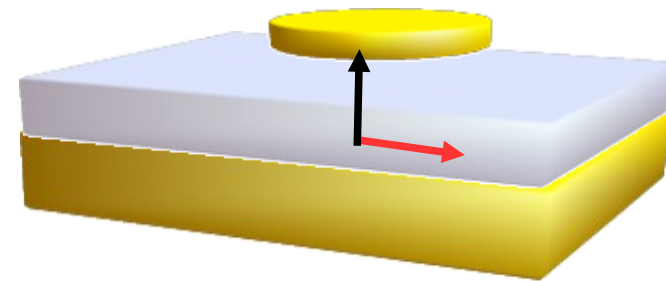
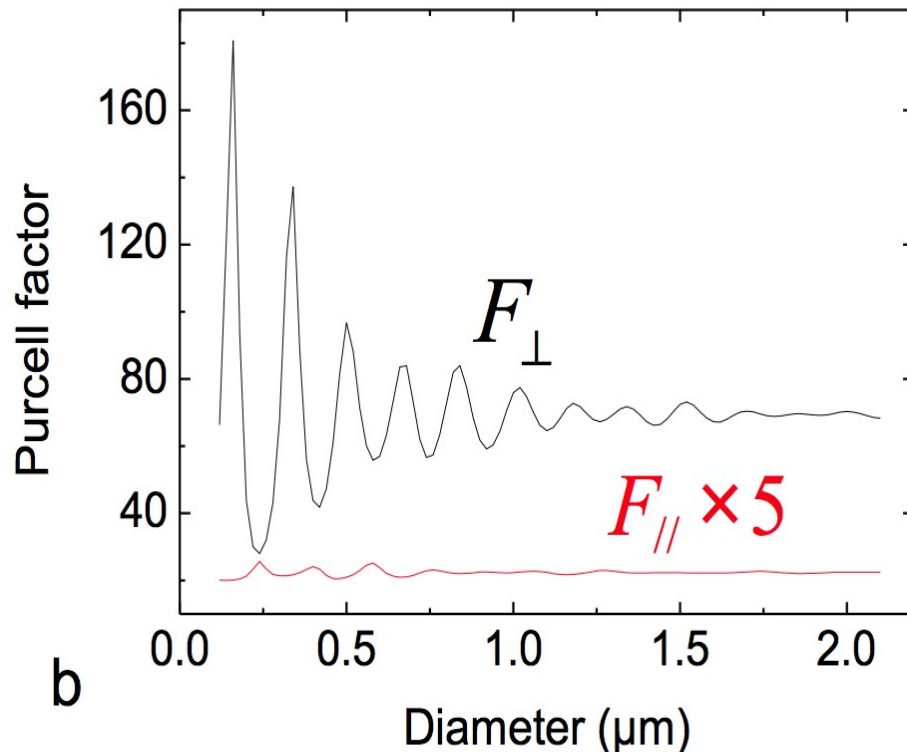
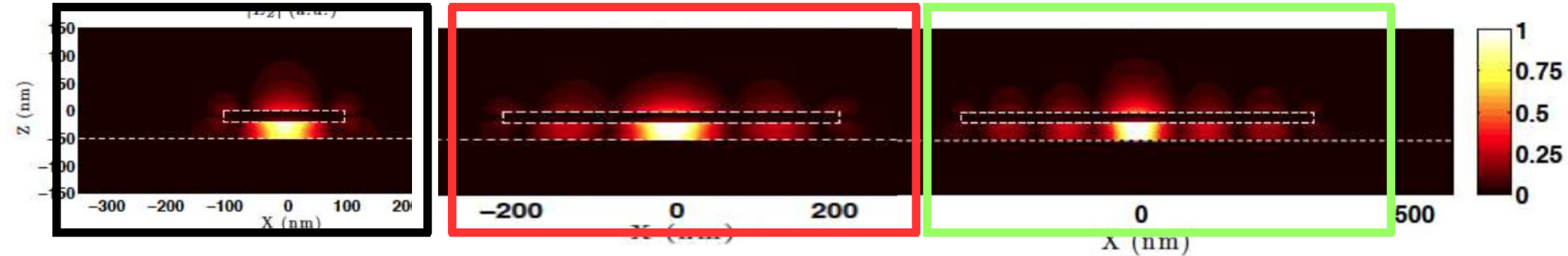
small antennas = high enhancement

The patch antenna: radiation pattern



Large antennas = better directivity

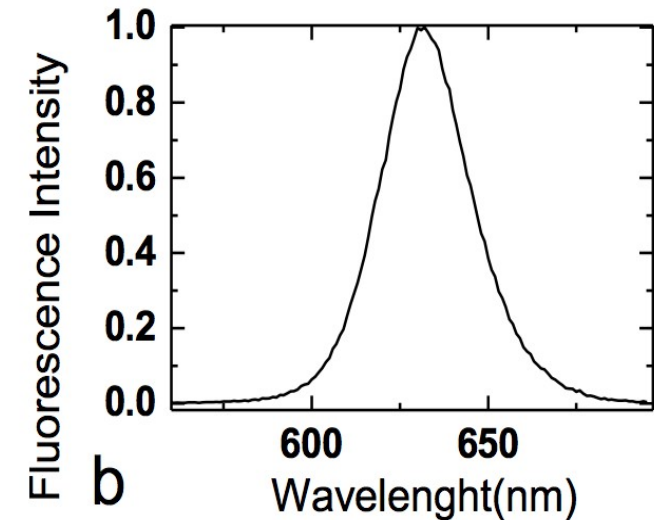
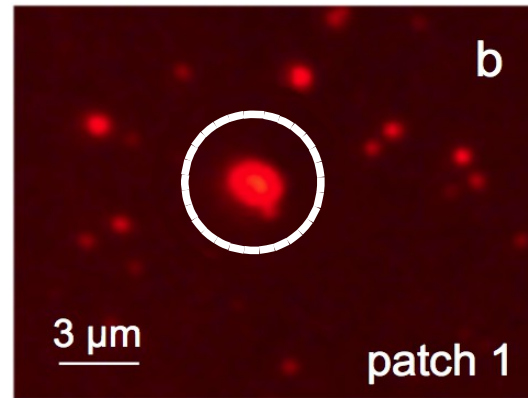
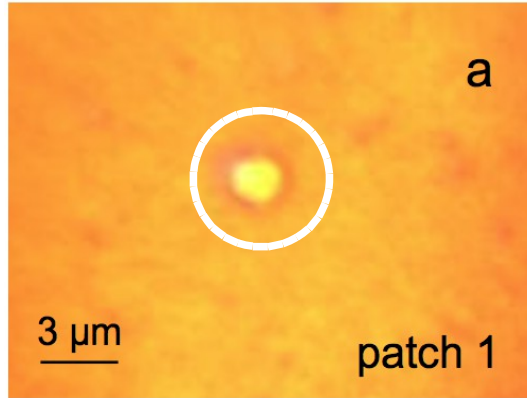
The patch antenna: dipole orientation



The purcell factor depends drastically on the dipole orientation
Cannot be controlled experimentally



The patch antenna : experimental results



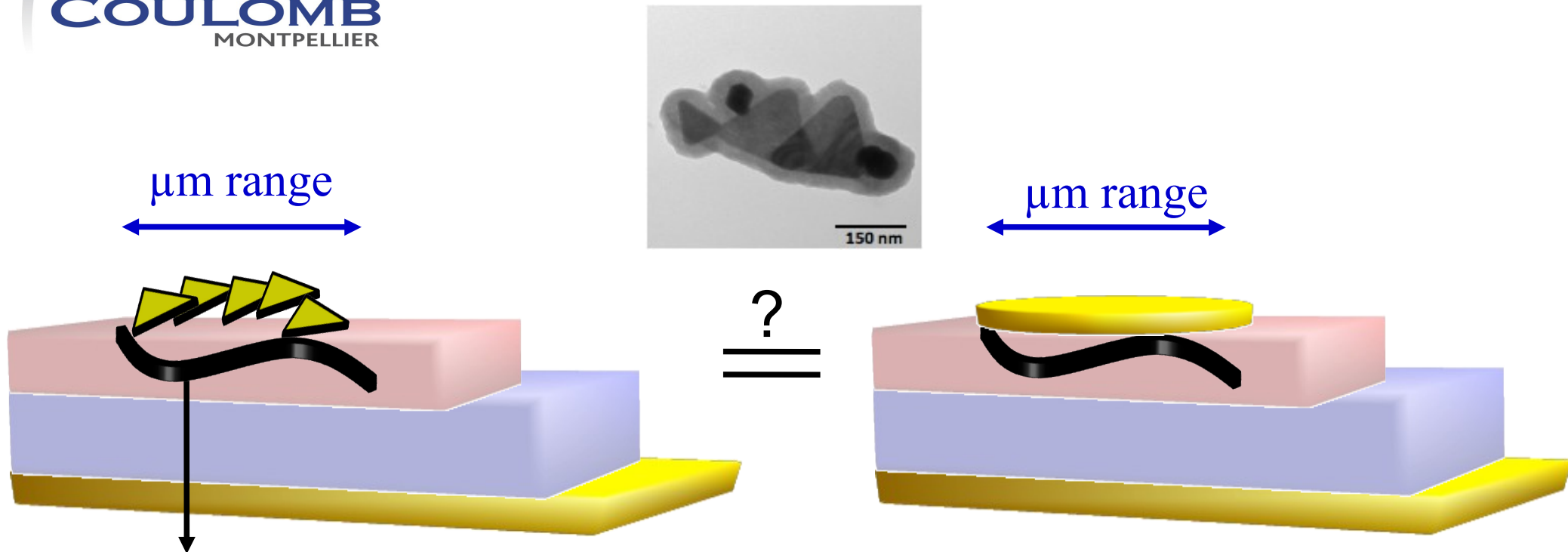
The whole metallic disk radiates
→ the plasmonic mode radiates

No modifications of
the emitter spectrum

Purcell factor ~ 70

Belacel *et al.* **Nano Lett.**, 2013, 13 (4), pp 1516–1521

plasmonics metasurface ?



Collection of individual scatterers

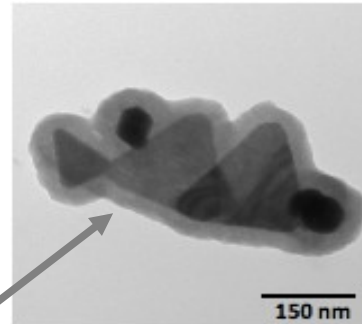
Advantages of both scale:

- Microscopic scale: dipolar resonance → Large Purcell factor
- large metallic structure → high directivity
- random distribution of plasmonic nanoparticles → relax dipole/metal surface alignment

Cluster of plasmonics nanoparticles

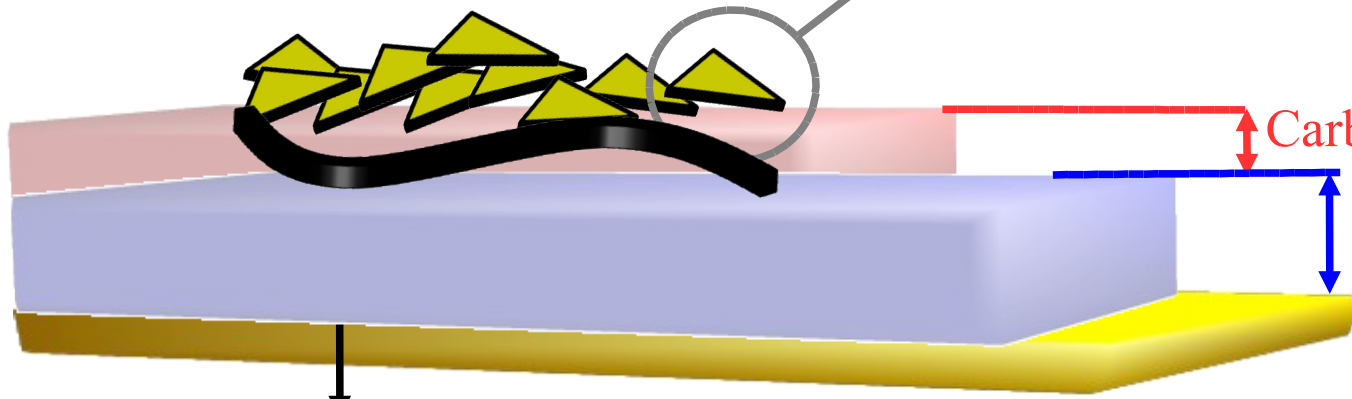
Incident light

μm range



Metallic nanoparticle resonant with the quantum emitter @ 1280 nm

Dipolar plasmonic mode



Carbon Nanotubes layer : 5 nm

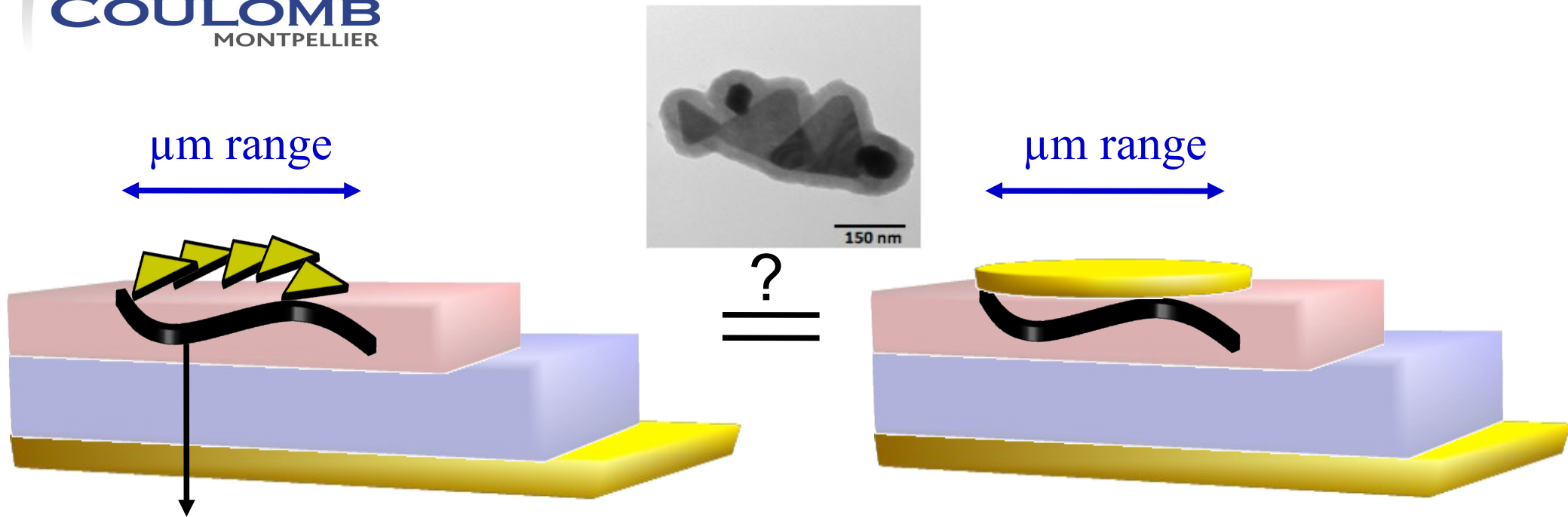
Dielectric spacer :
from 0 to 10 nm

Single emitter = Carbon Nanotube

Collective response mediated by the metallic plane ?

Surface Plasmon propagates on 100's μm range in the visible (in the near-IR)

plasmonics metasurface ?



Collection of individual scatterers

Experimental point of view:

Conditions to get a collective response ?

Cannot distinguished the contribution of one specific nanoparticle, contribution of one specific carbon nanotube

=> the whole cluster radiates

=> With the same spectrum

The experimental setup:

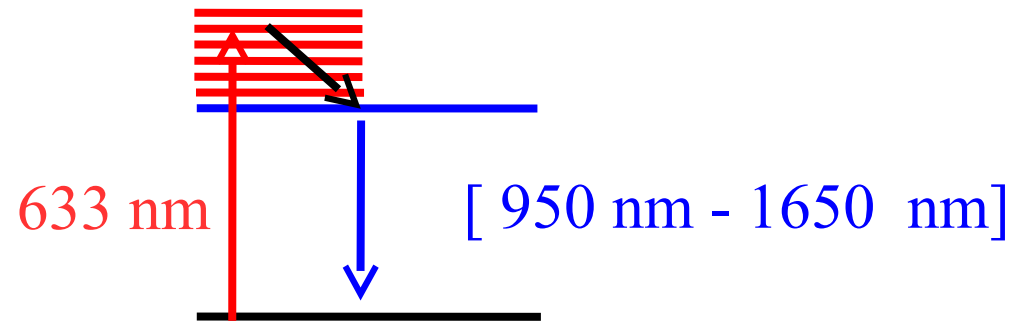
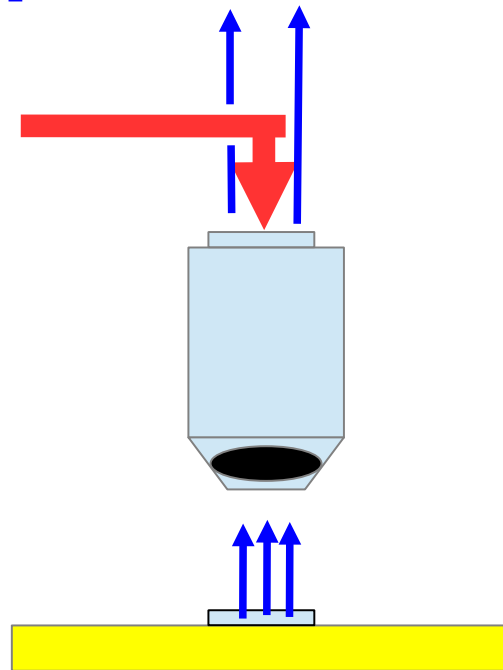
Luminescence spectrum

Spectrometer :

Luminescence spectrum
[950 nm - 1650 nm]

In the near-IR window
Long-distance propagation length
of the surface plasmon

Excitation laser
@ 633 nm



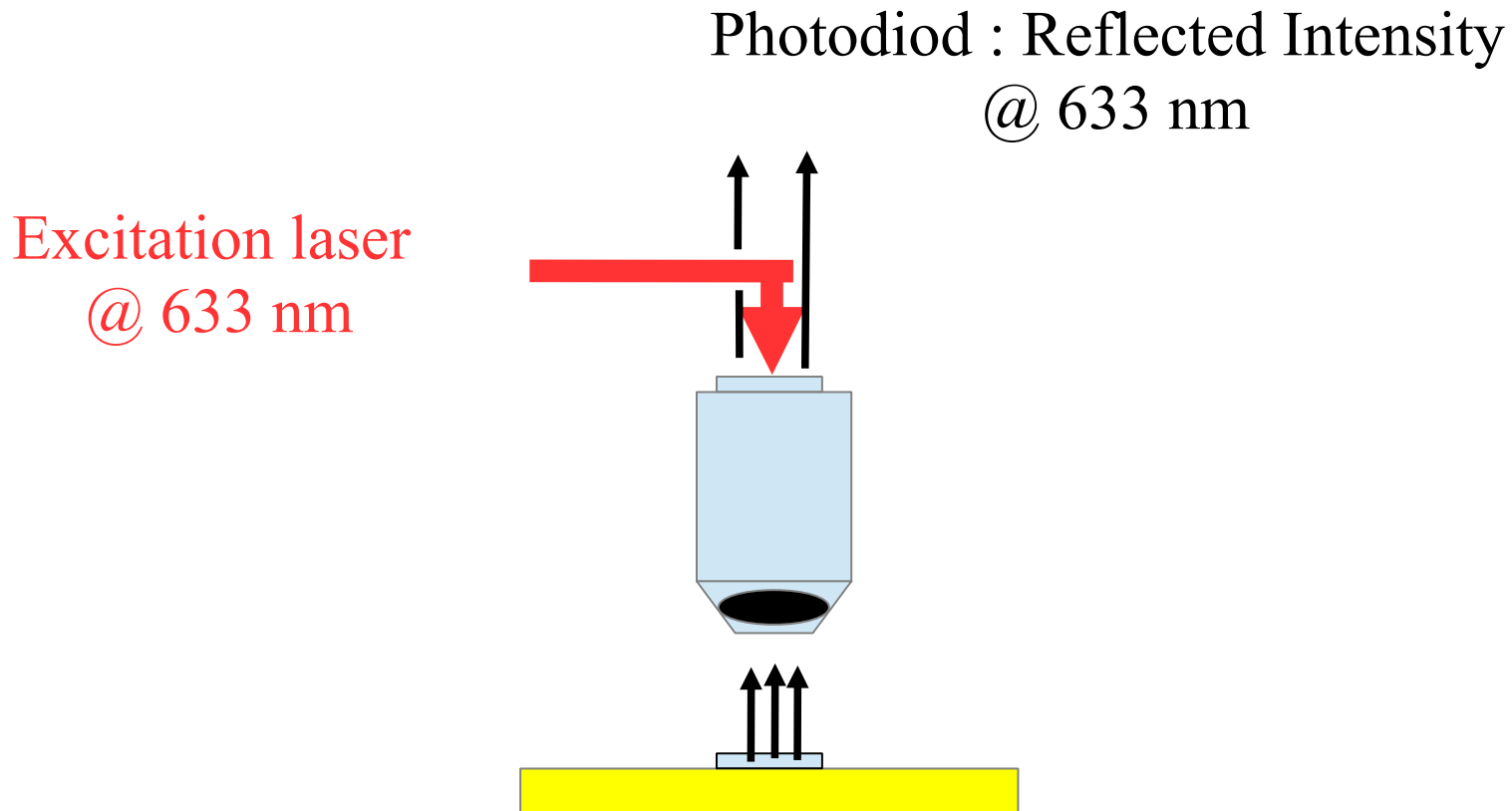
Cartography of the sample luminescence (XY-
map)

From room temperature to 4 K

emmanuel.rousseau@umontpellier.fr

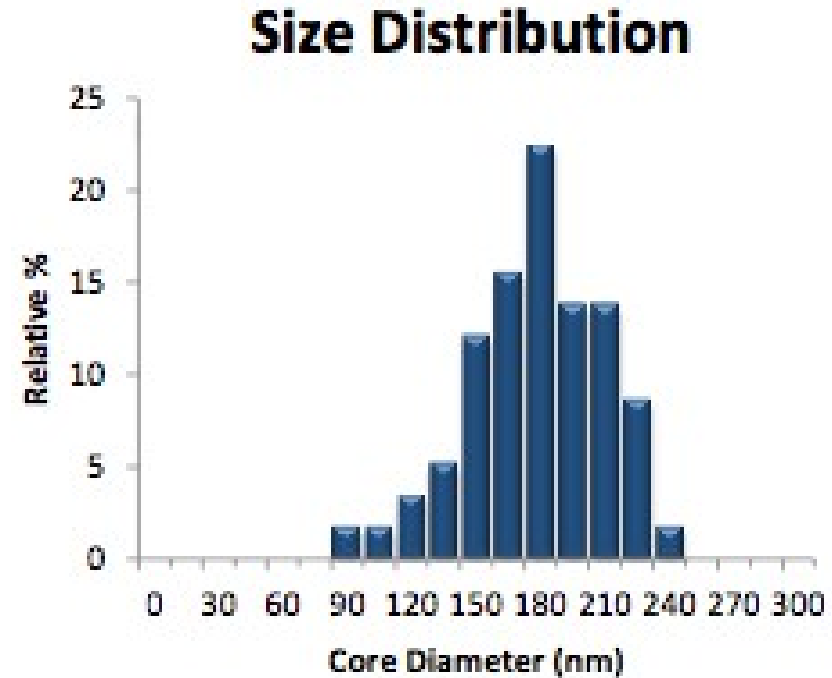
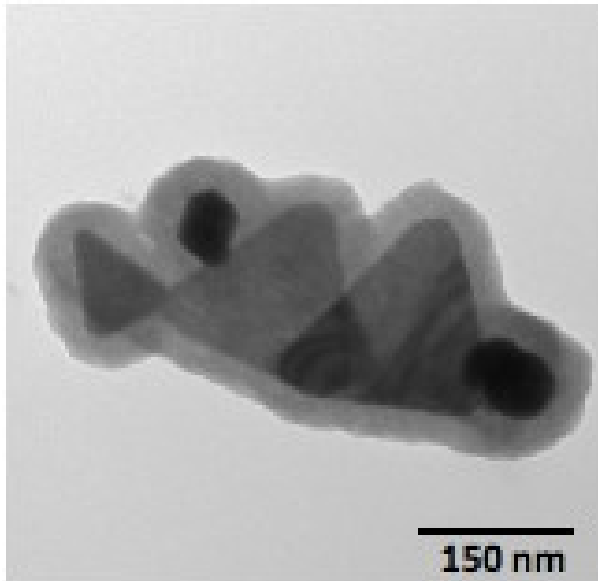
The experimental setup:

Reflectivity



Cartography of reflectivity of the sample surface (XY-map)

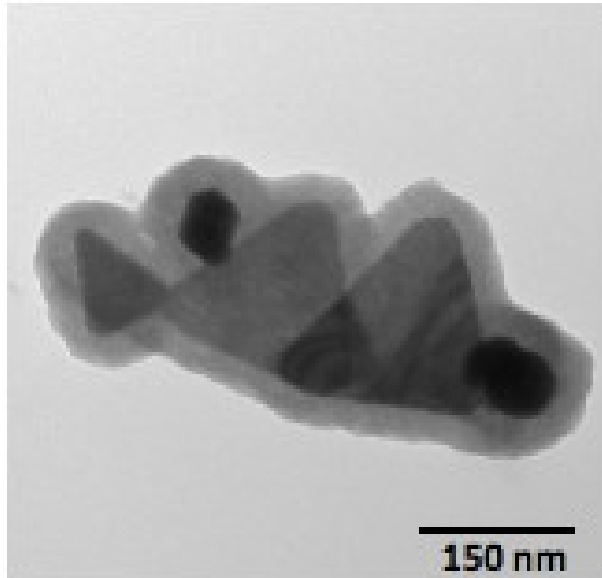
The sample : the plasmonic nanoparticles



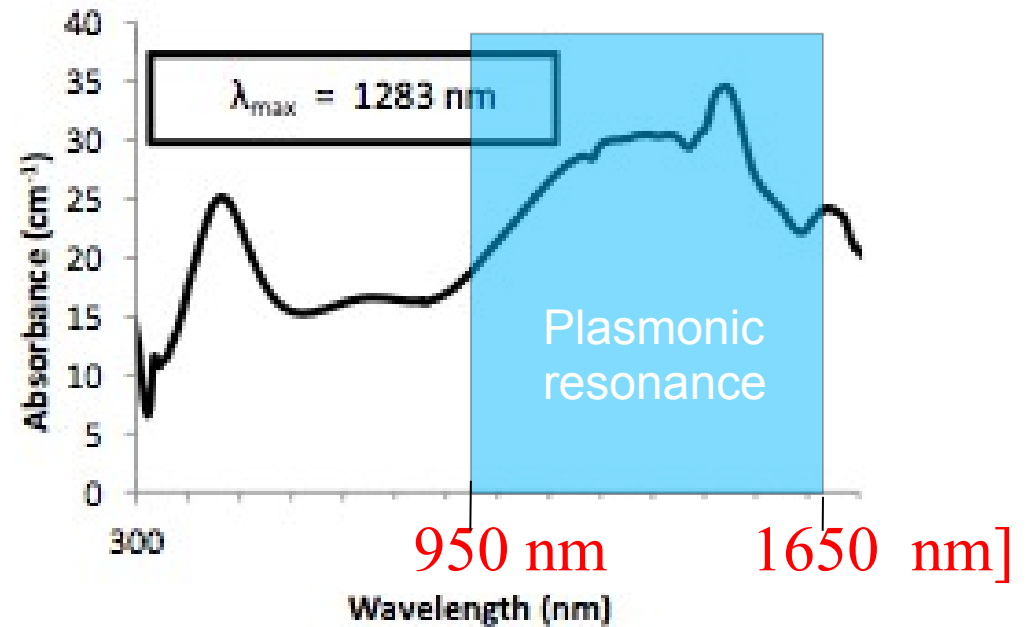
Patch: Colloidal nano-triangles made on silver coated with silica

Resonant @ 1280 nm
with broad size dispersion

The sample : the plasmonic nanoparticles



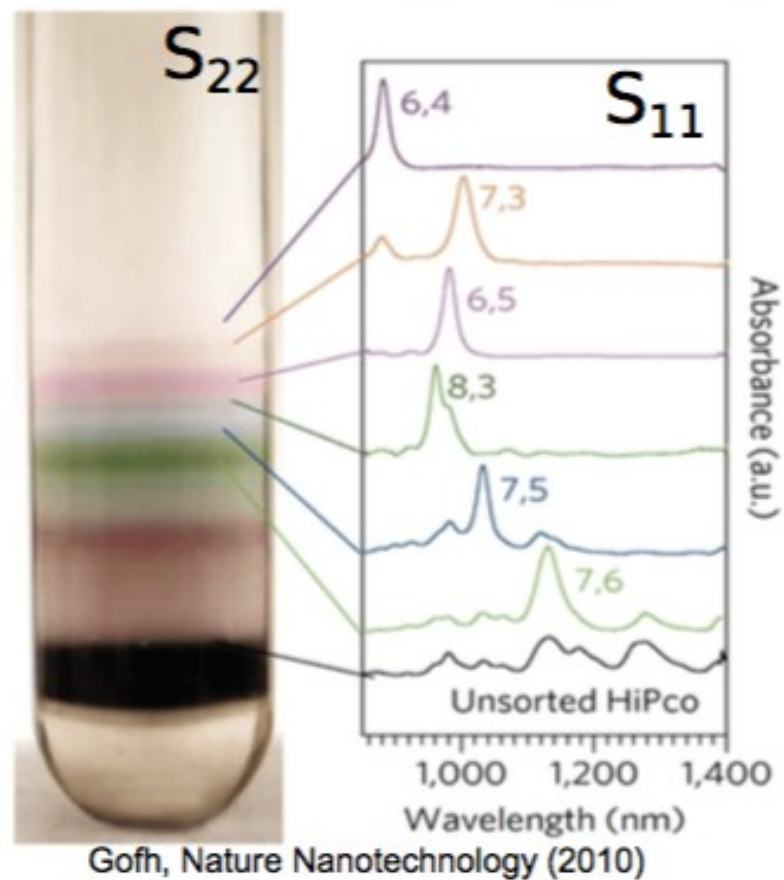
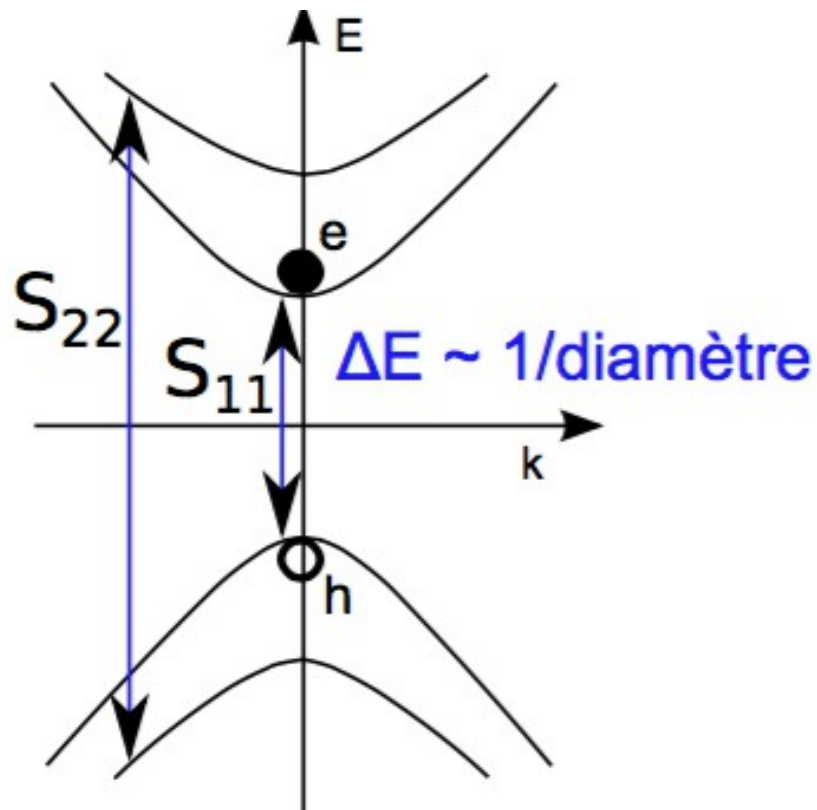
Optical Properties



broad size dispersion => broad “resonance” peak
inhomogeneous broadening



The sample : the carbon nanotubes

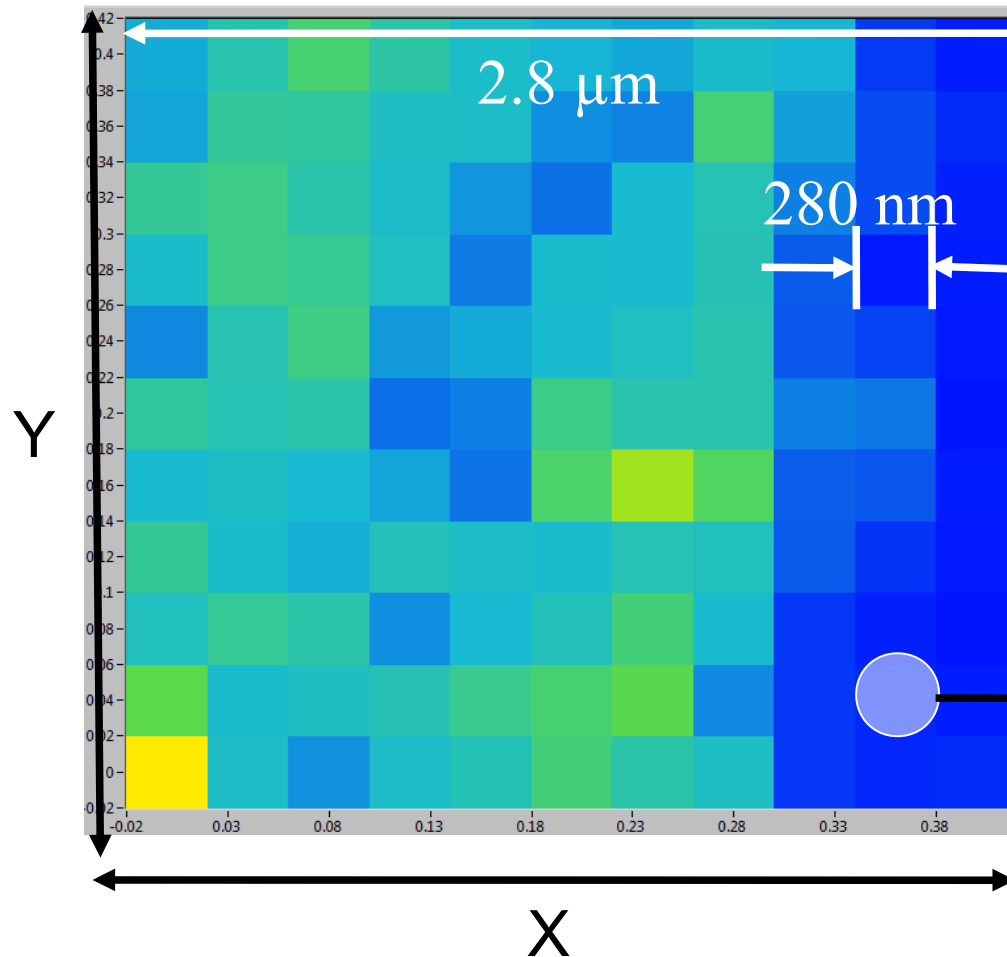
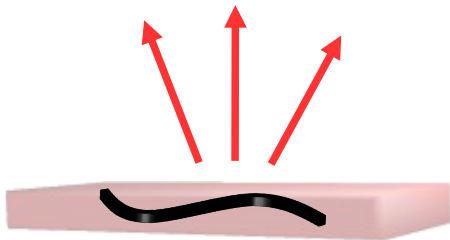


Typically : 5 families of semiconducting carbon nanotubes

Map of the sample luminescence

XY-Map of the Total Intensity

Nanotube
luminescence

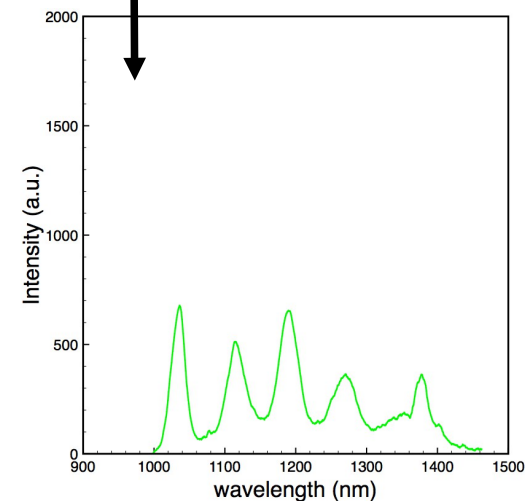
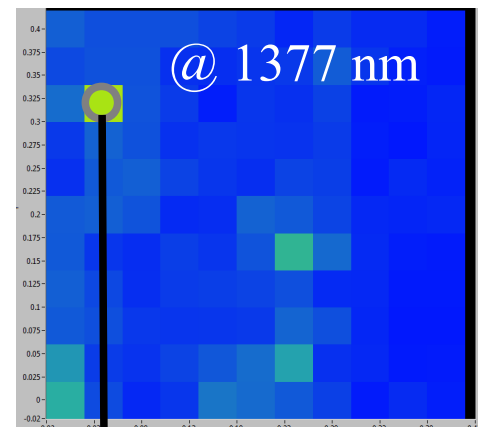
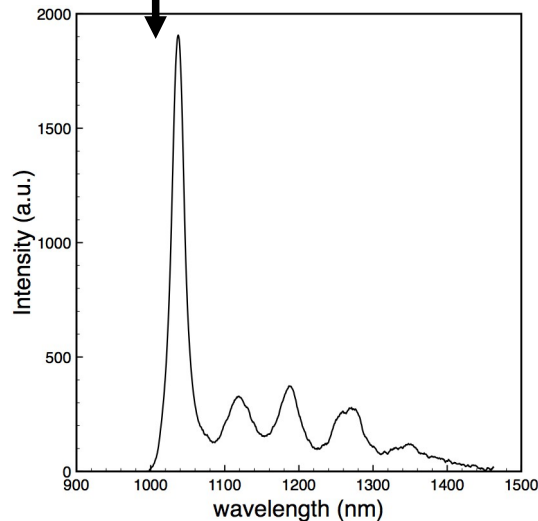
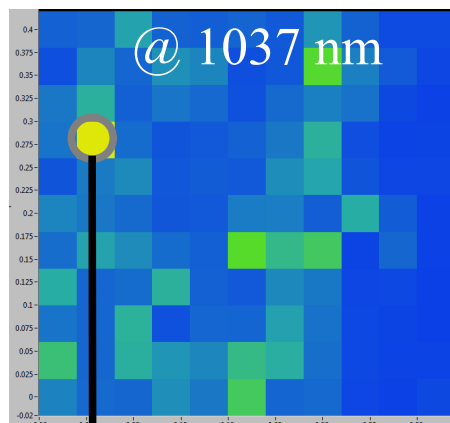
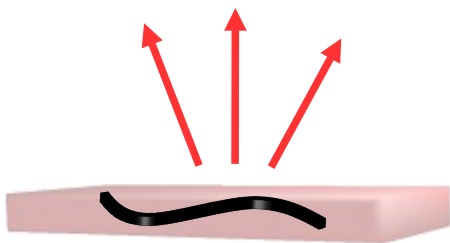


Excitation
laser Spot size

~
Step size

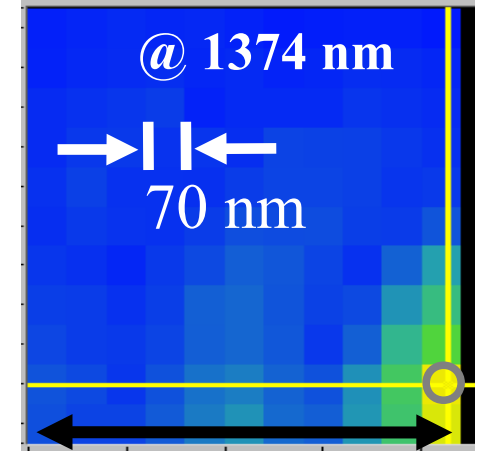
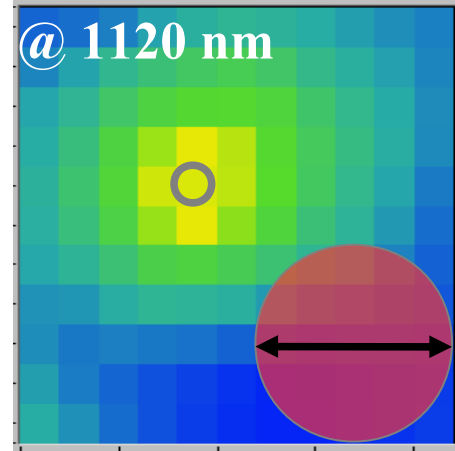
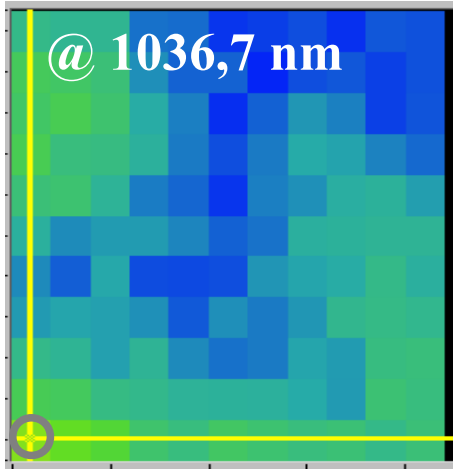
Map of the sample luminescence: spectral variations

Nanotube
luminescence



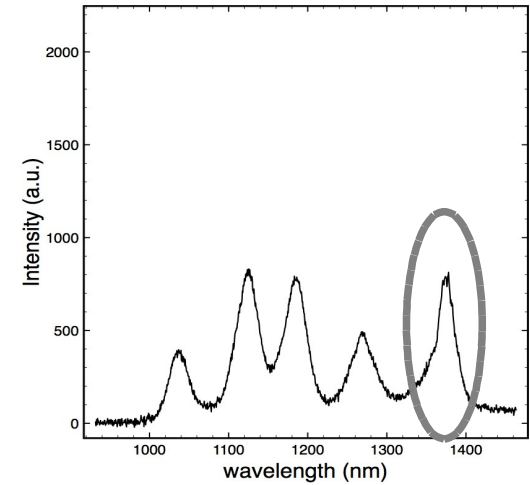
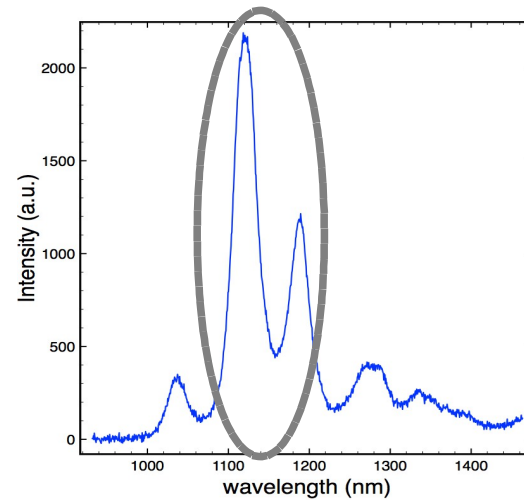
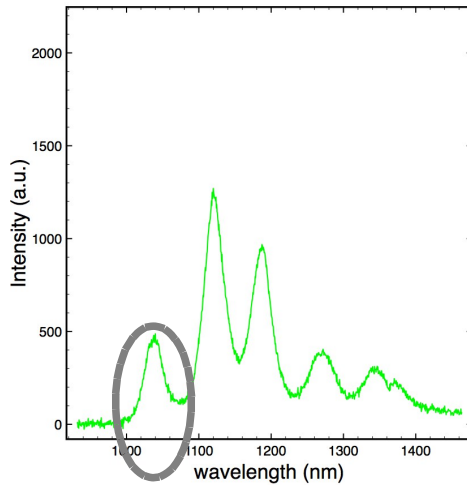
Typically : 5 families of carbon nanotubes with well-defined peak

Cartography of the sample : finer scale



Excitation laser Spot size

700 nm

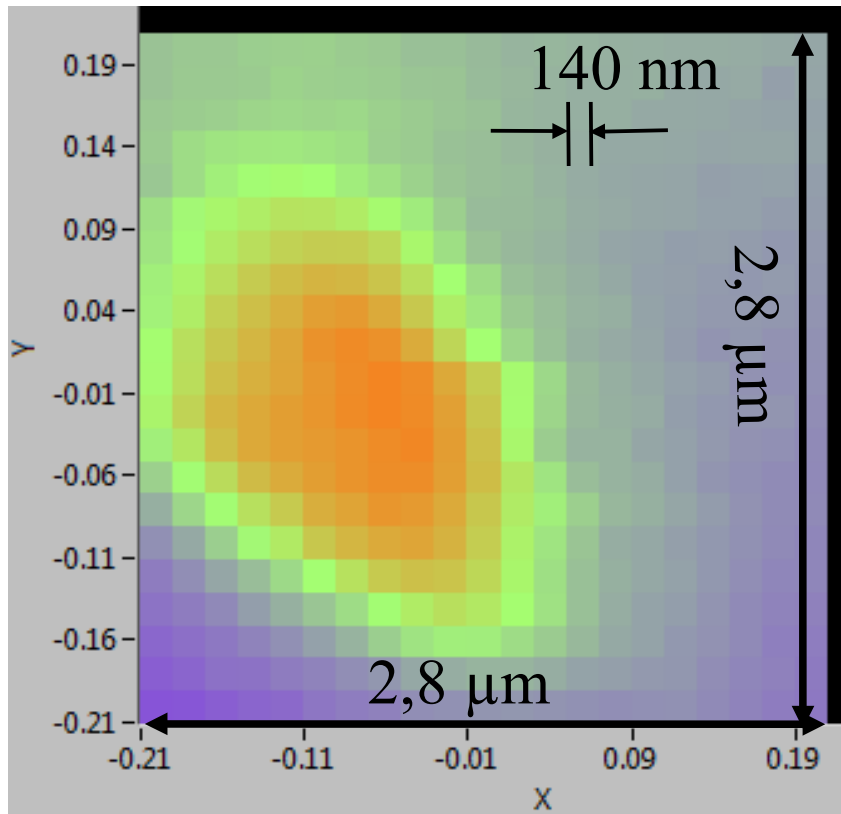


Inhomogeneity characteristic scale ~ 500 nm

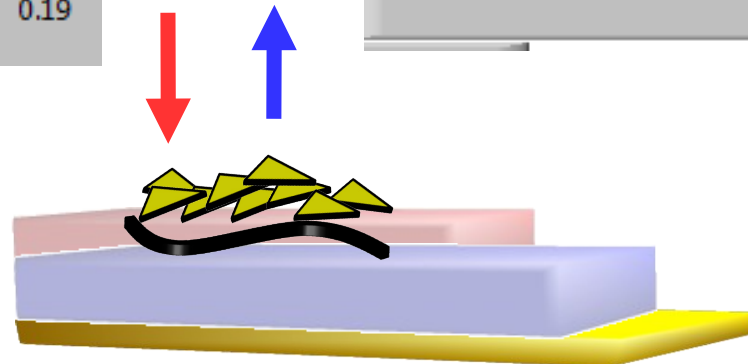
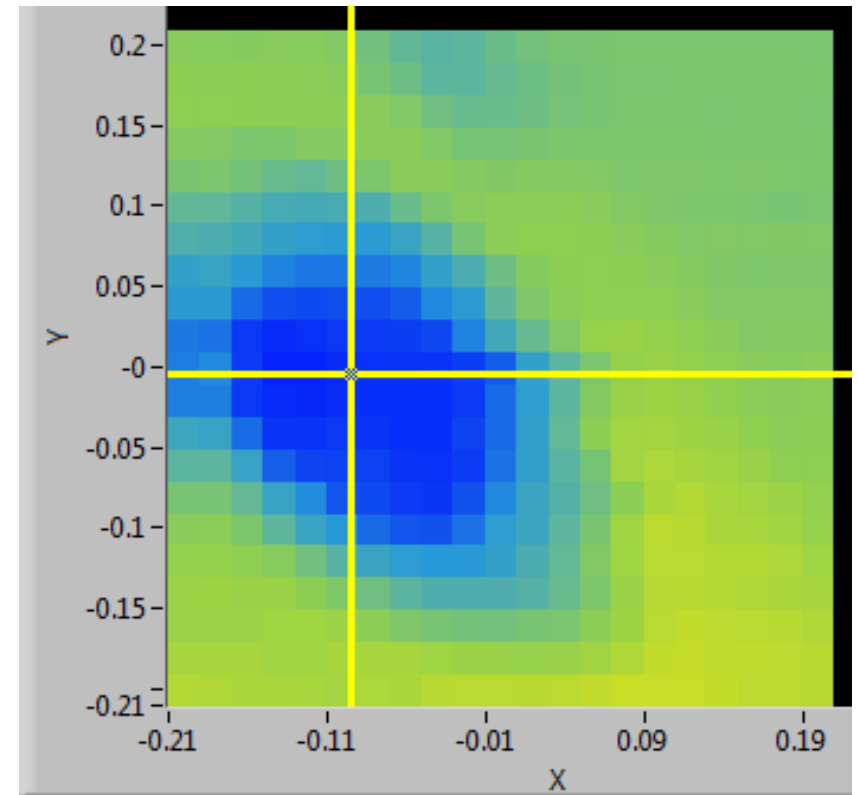


Plasmonics and plasmonics antennas

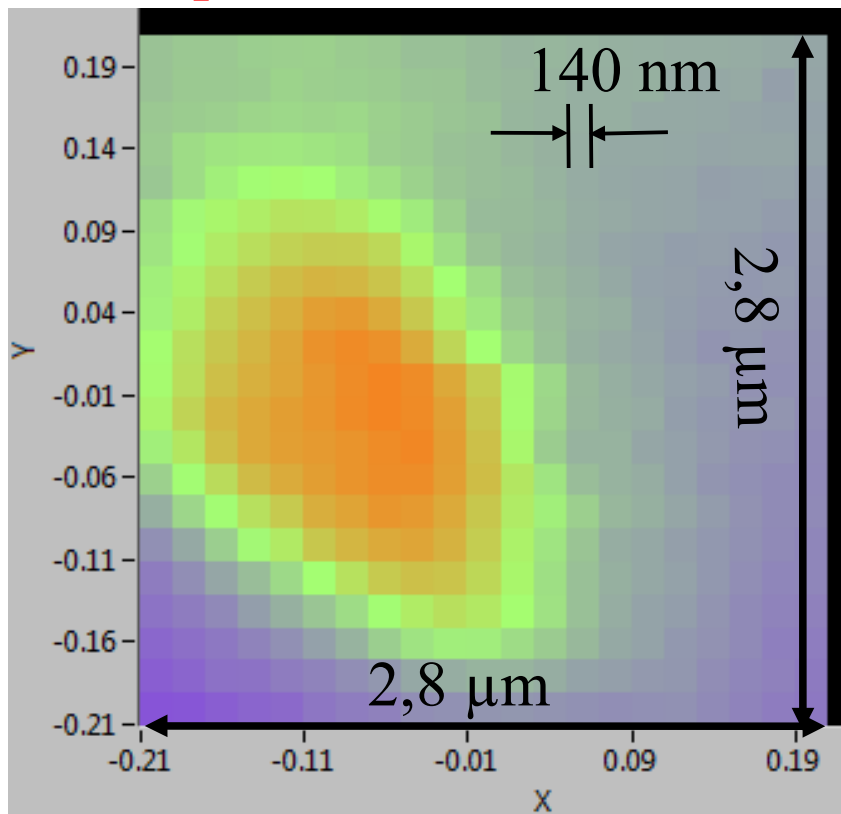
photoluminescence



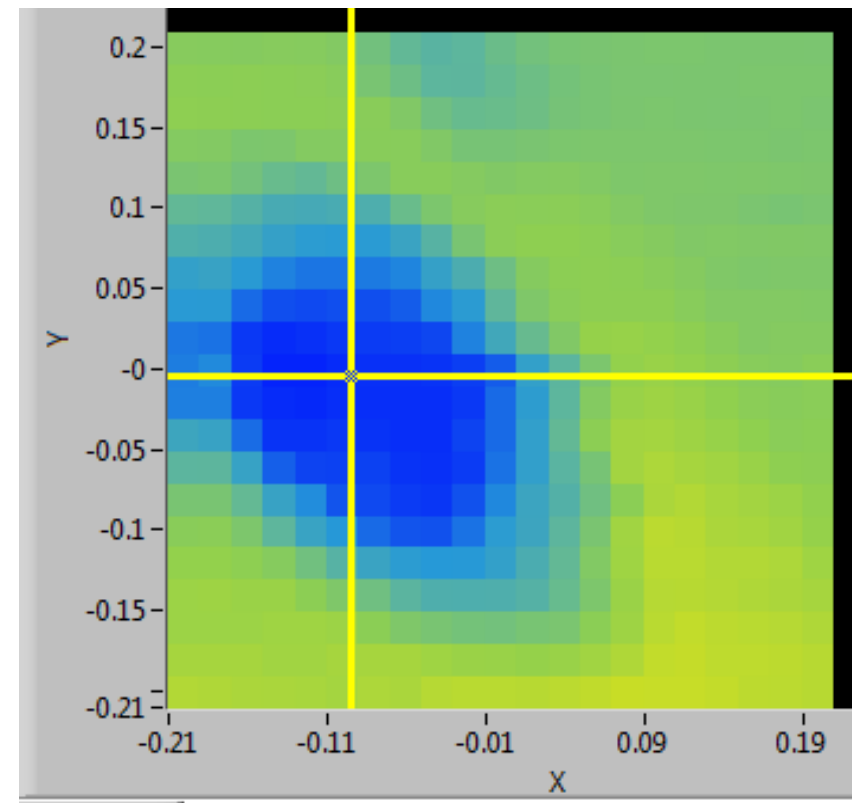
Reflectivity



photoluminescence



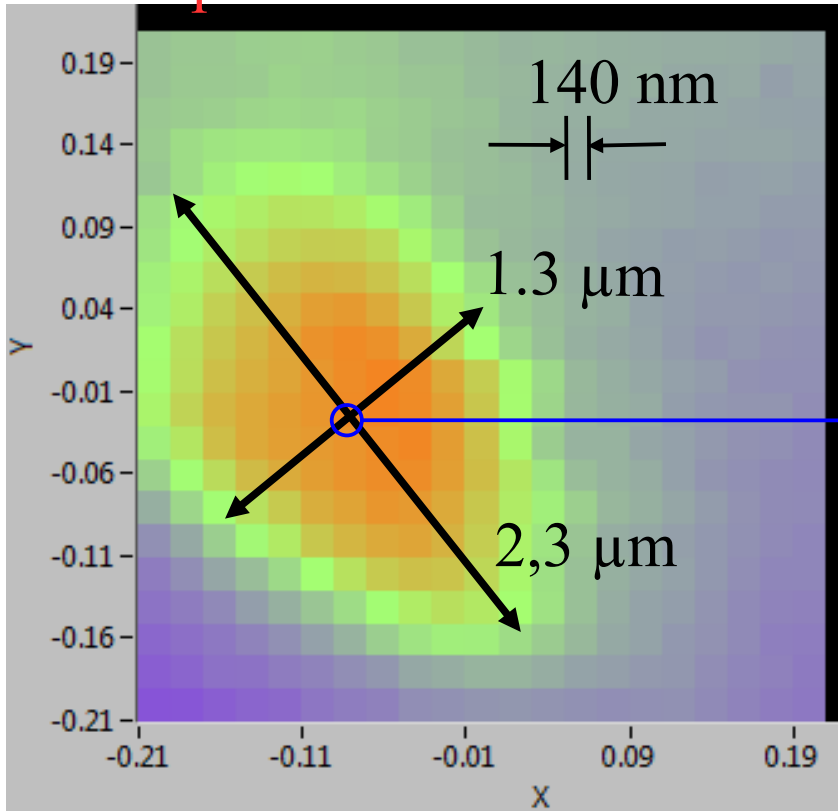
Reflectivity



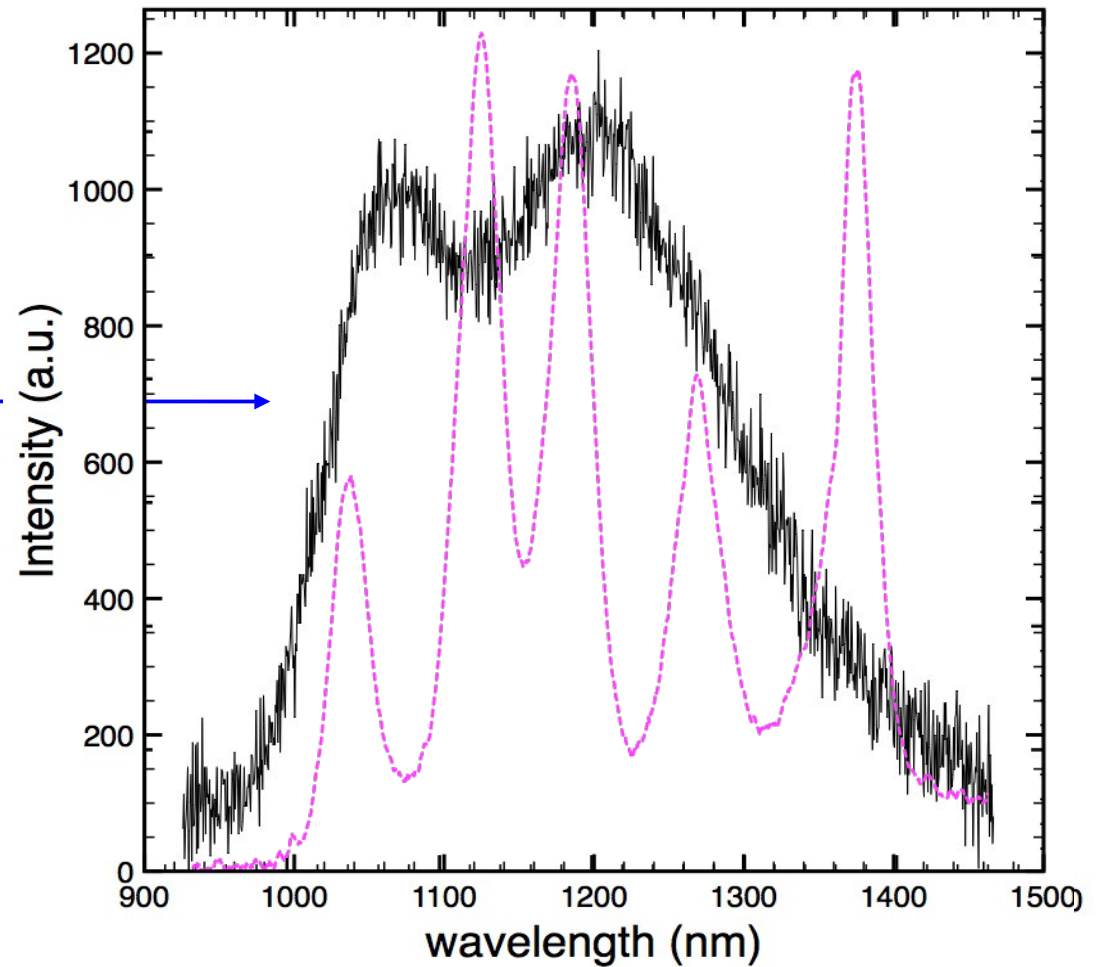
Exact match between luminescence map and reflectivity map
The nanoparticles cluster enhance the luminescence

Plasmonics and plasmonics antennas

photoluminescence

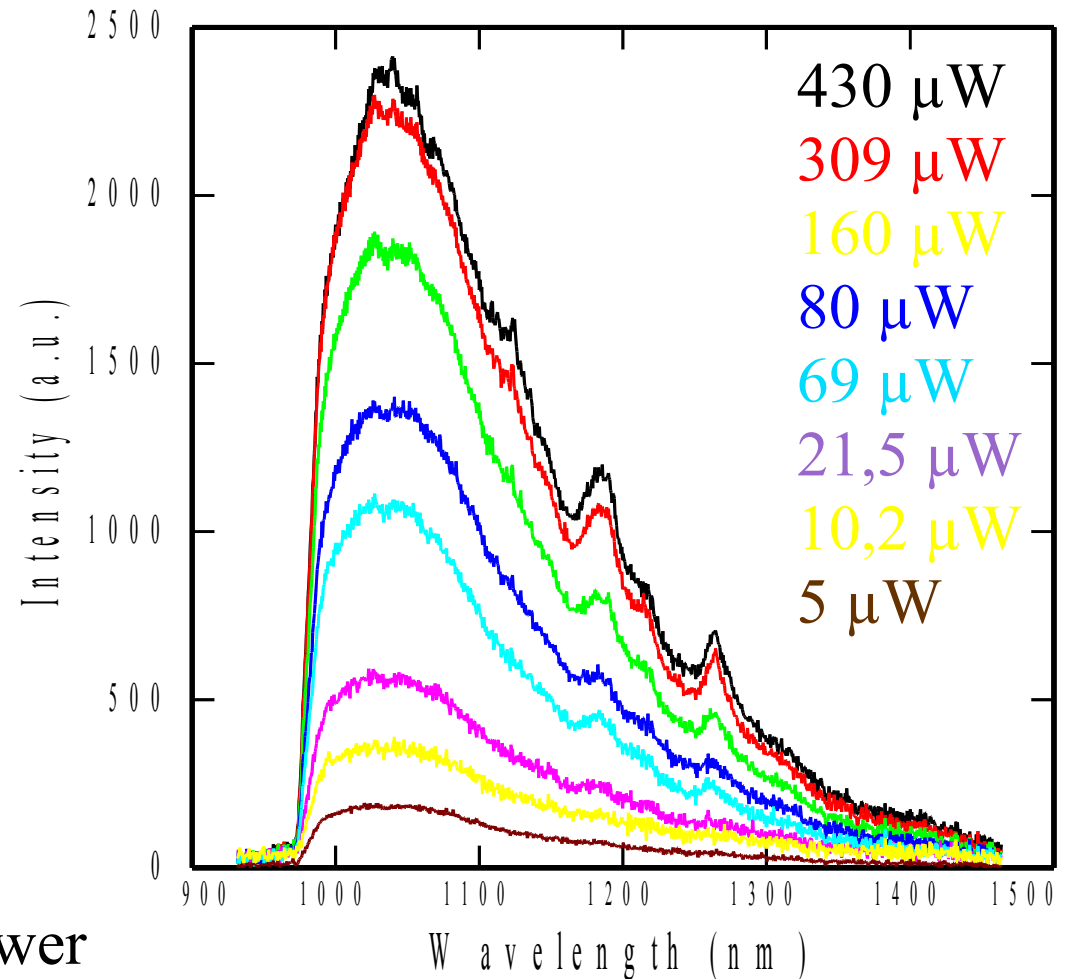
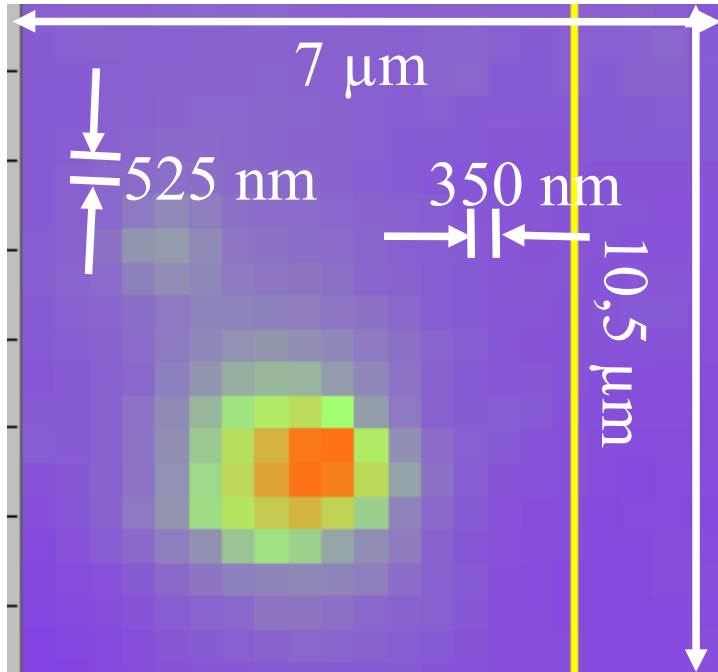


Spectrum



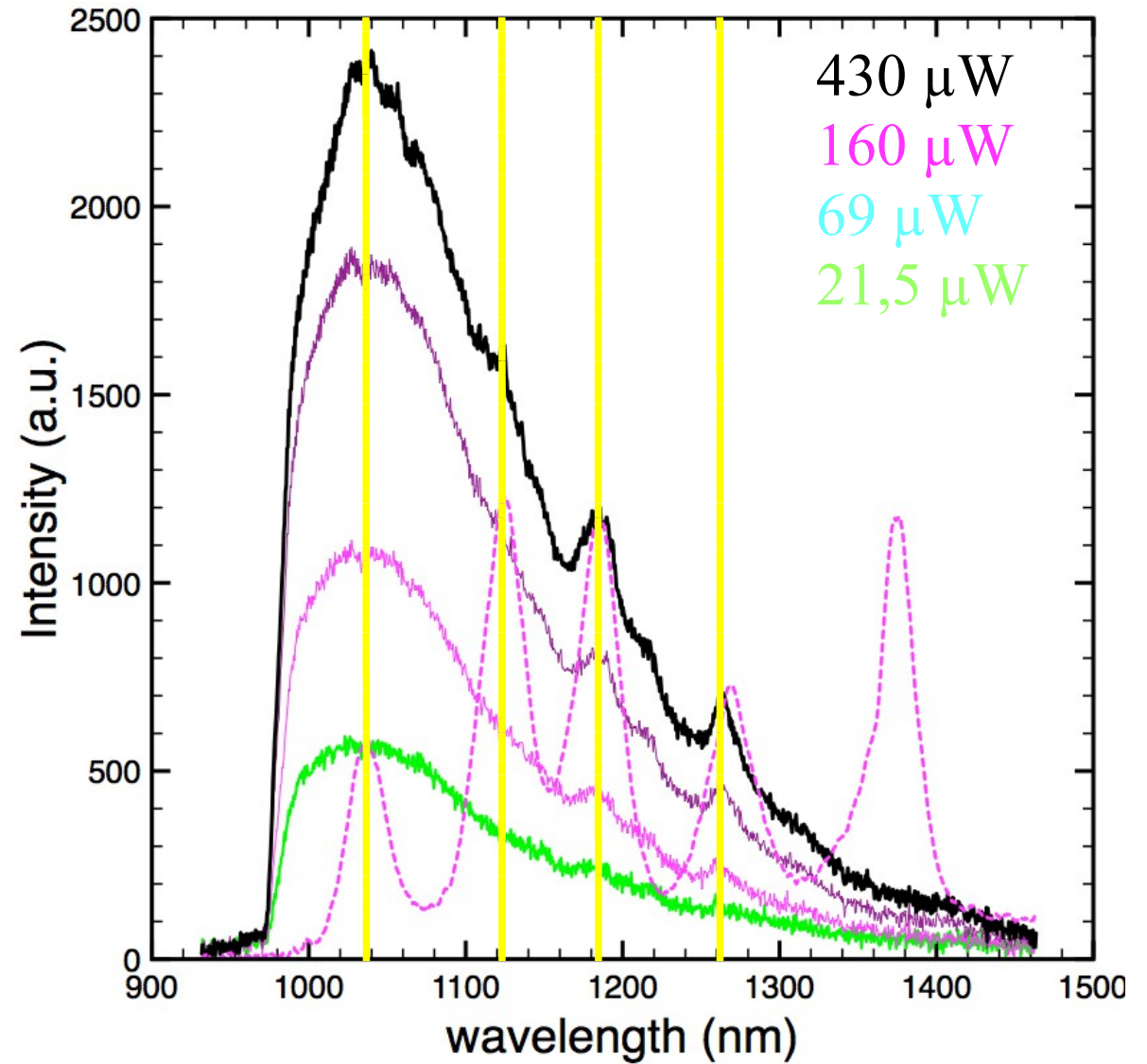
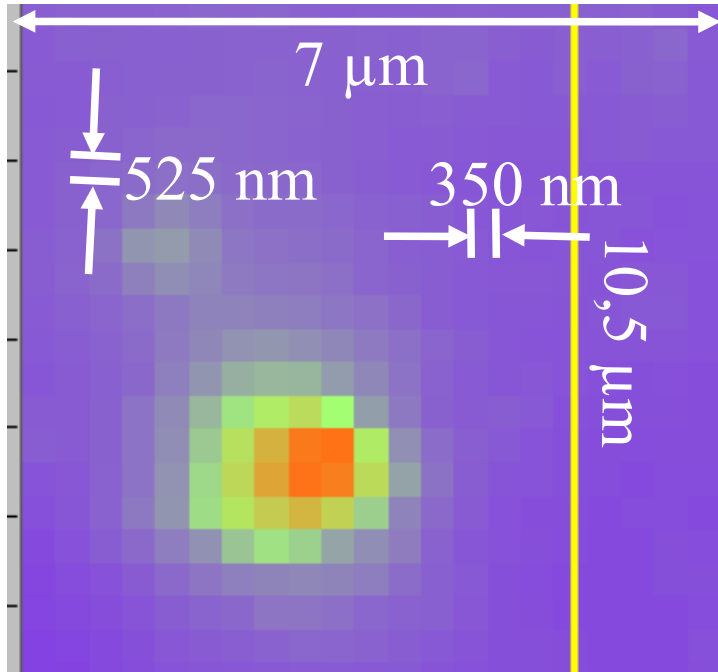
The spectrum is broad

Spectrum shape is similar on scale $\sim 2 \mu\text{m}$

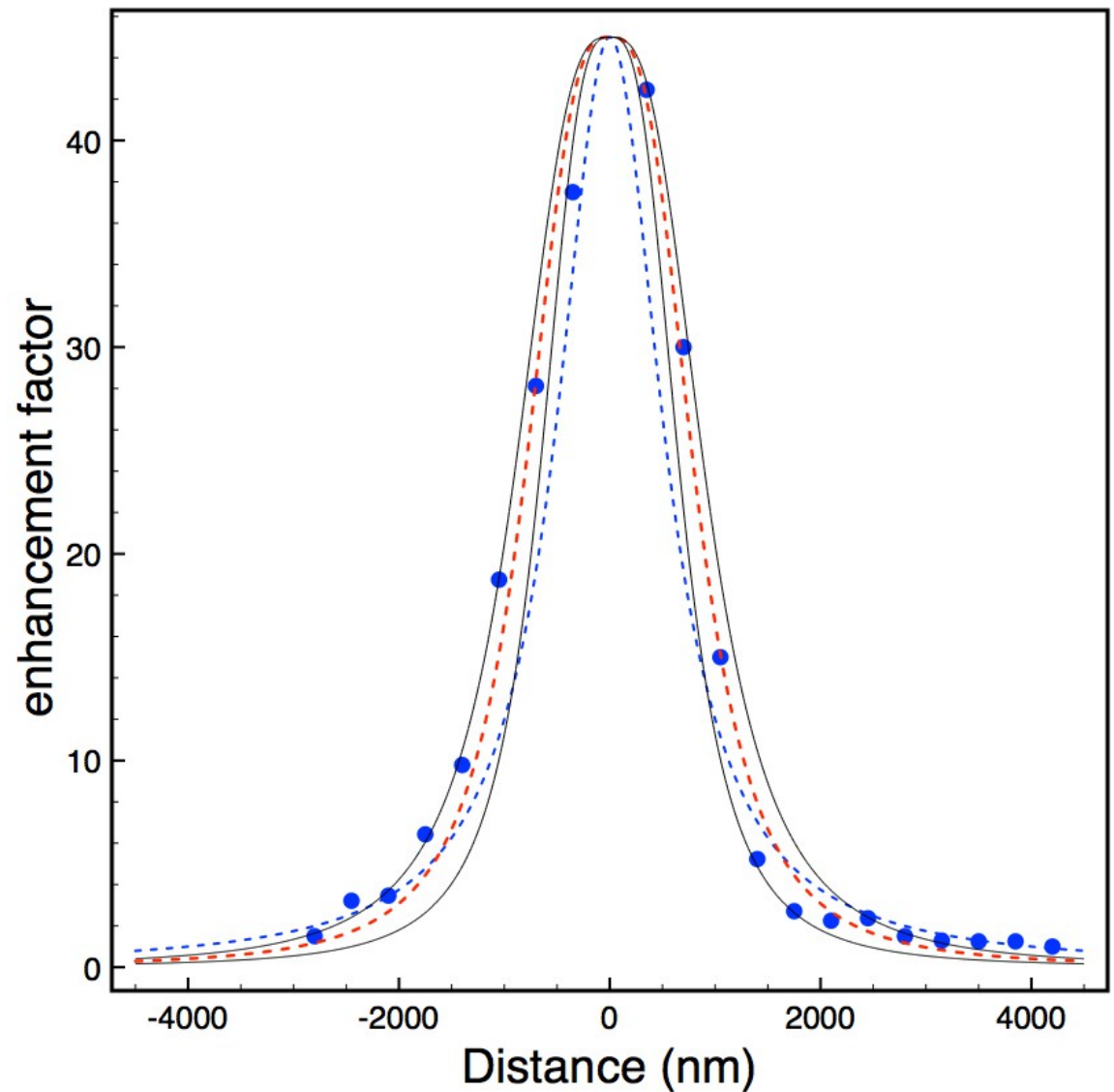
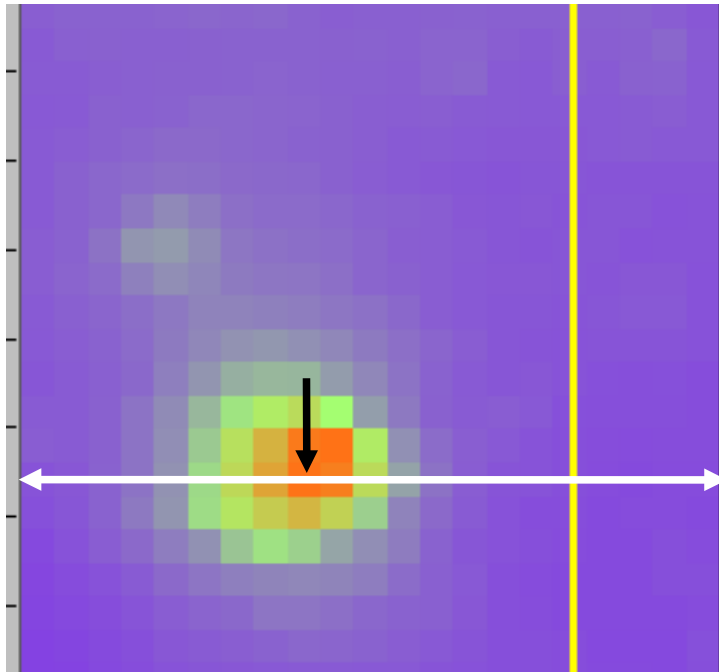


When increasing excitation power

NT characteristic peaks (re)-appears !

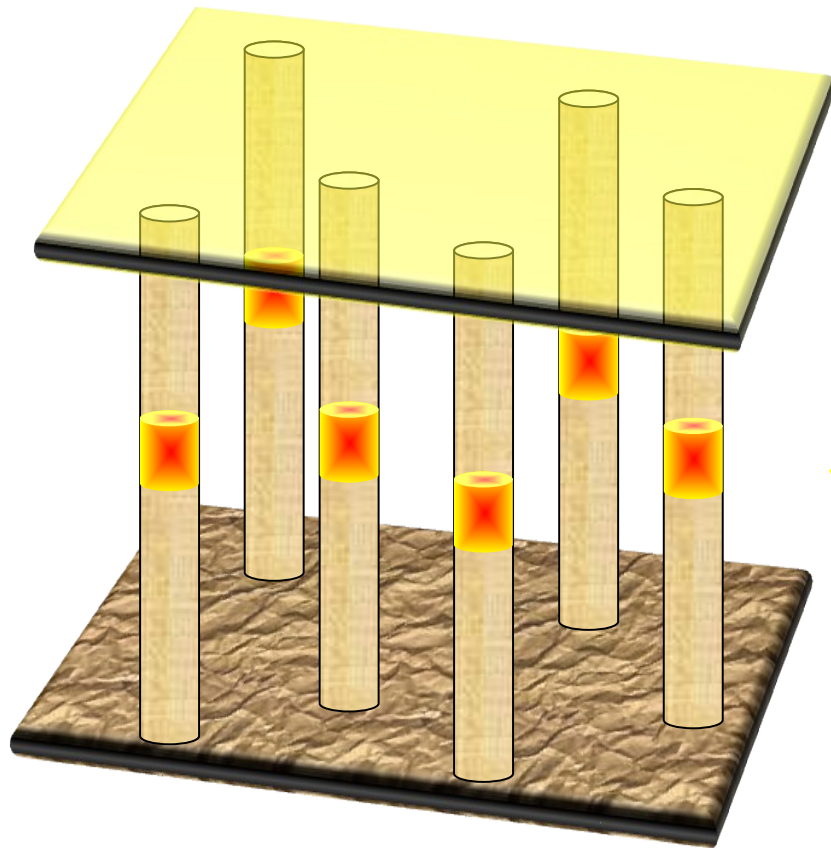


Enhancement of the luminescence



Enhancement of the luminescence by a factor ~ 45

- Collective behavior for a cluster of plasmonic nanoparticles
 - *cannot distinguish the contribution of a single nanoparticle, on single carbon nanotubes*
 - *plasmonics metasurface?*
- radiative enhancement ~ 45
- Spectrum broadening ? → from 4K to 300 K !
 - Why CNT peaks reappear when increasing power?
 - failure of the two-levels model
- *emission diagrams measurements?*
- *numerical simulations (disorder, frequency mismatch).*



Dielectric cylinders:

- periodic shaping (or not)

Quantum dots

=> How Quantum dots modify
light propagation?

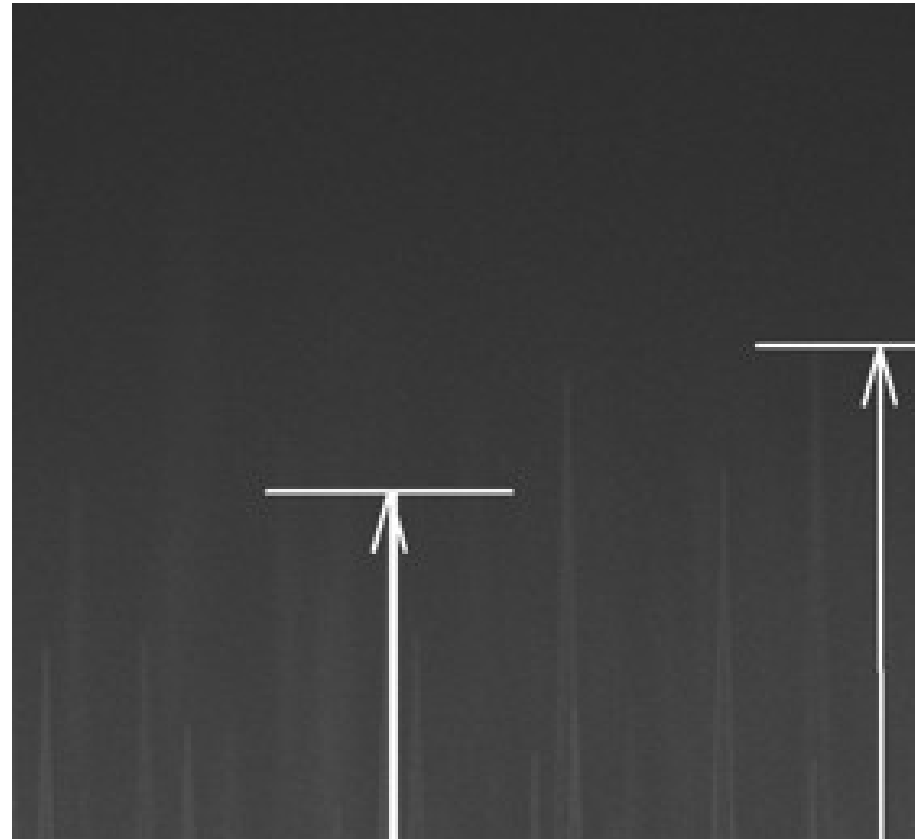
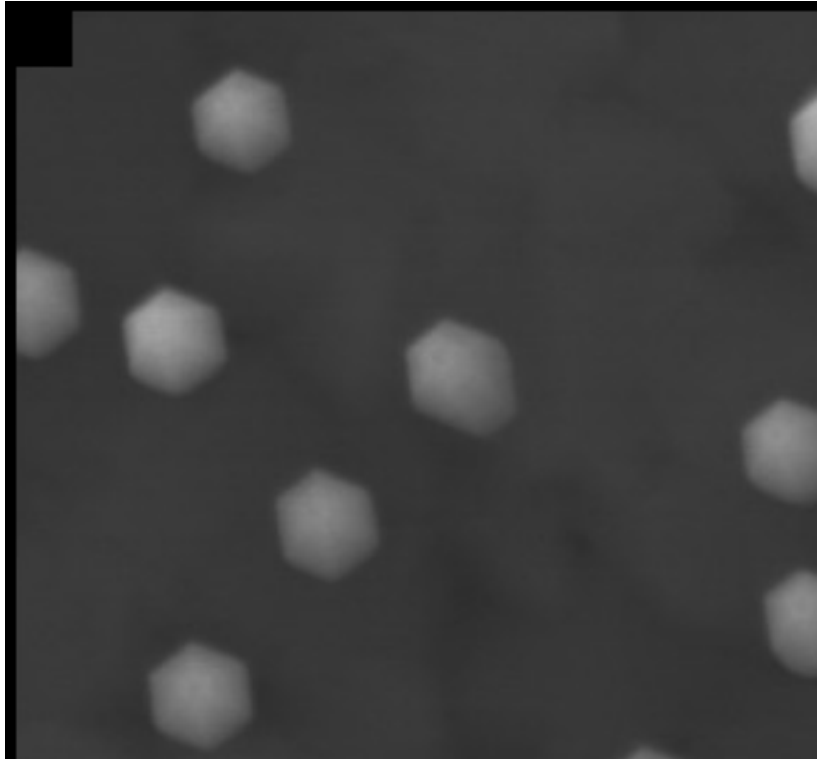
Simplifications:

2D geometry

spatial confinement

Nanorods: InP refractive index ~ 3.17 @ $1.5 \mu\text{m}$

Quantum dots: InAsP the emitting wavelength is in the near-IR $1.5 \mu\text{m}$



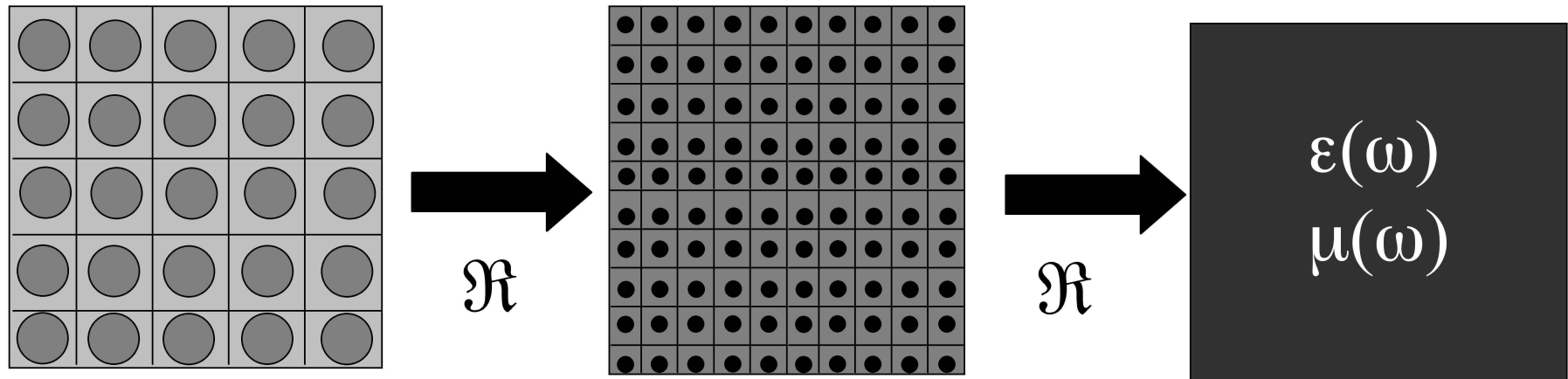
Diameters: $\sim 100 \text{ nm} \pm 10 \text{ nm}$ but can be adjusted from 50 nm to 500 nm during growth

Length: $\sim 3 - 4 \mu\text{m}$ up to $10 \mu\text{m}$

Defaults free = very good optical properties

Encapsulated in resist, top metallic mirror

Homogeneization theory



The renormalisation transform is defined by:

size divided by η

Permittivity multiply by η^2

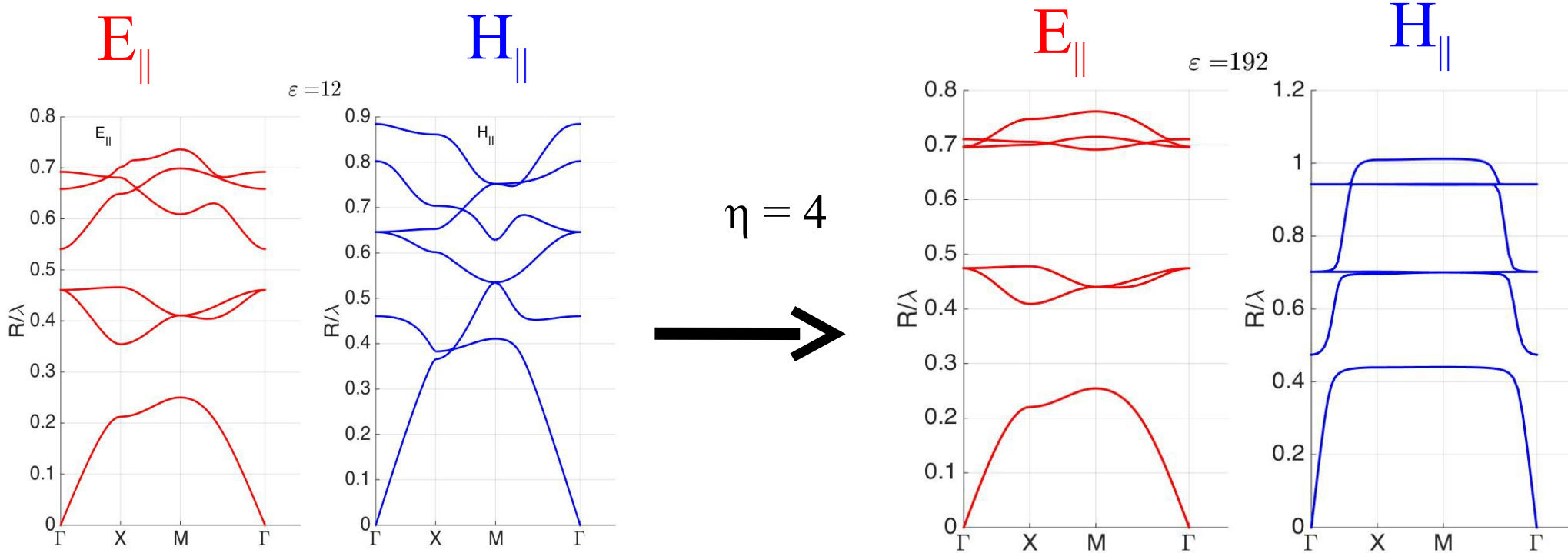
Wavelength been constant



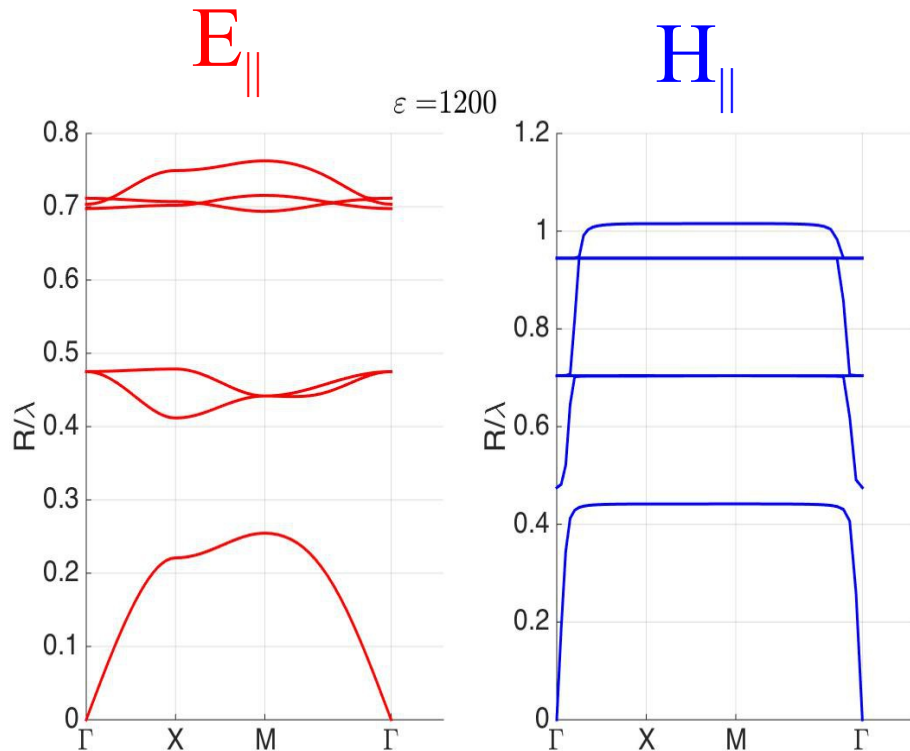
Constant optical diameter

How does this transform modify the band structure ?

Bloch diagrams



Bloch diagrams



The E_{\parallel} Bloch spectrum is unchanged by the renormalisation transform

Is there a homogenization result away from Γ ?

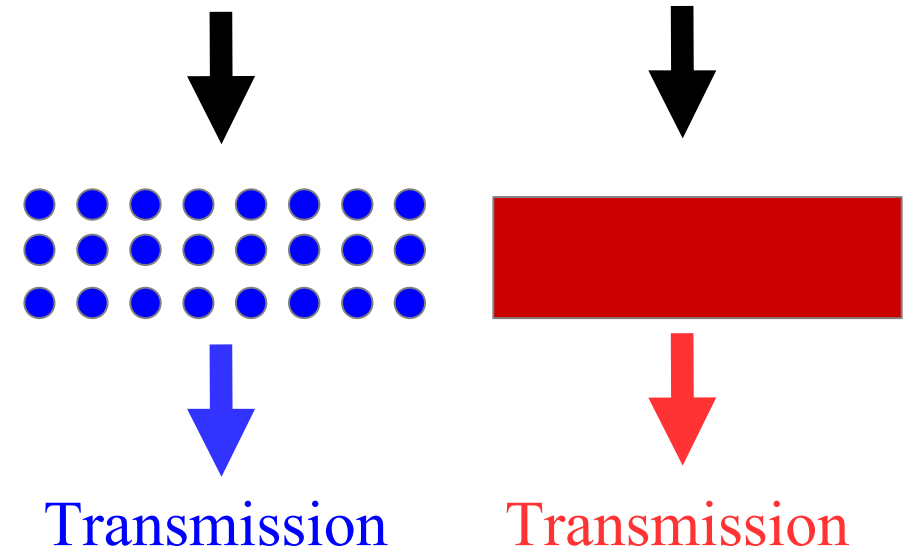
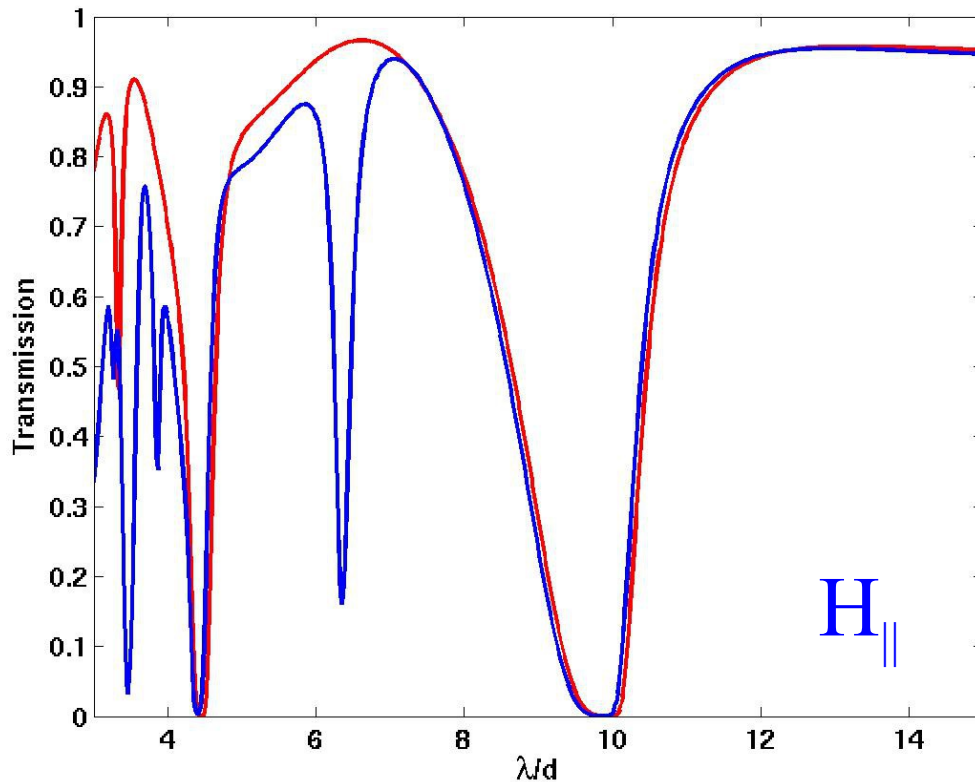
K.Vynck, D. Felbacq, Phys. Rev. Lett. **102**, 133901 (2009)

The H_{\parallel} Bloch spectrum converges towards that of a homogeneous medium with resonances

D. Felbacq, G. Bouchitté, Phys. Rev. Lett. **94**, 183902 (2005)

$$\mu_{\text{eff}}(\omega) = 1 + \sum_n \frac{\omega^2}{\lambda_n \epsilon_i^{-1} c^2 - \omega^2} \left(\int_D \phi_n(y) dy \right)^2$$

Light transmission through the rods



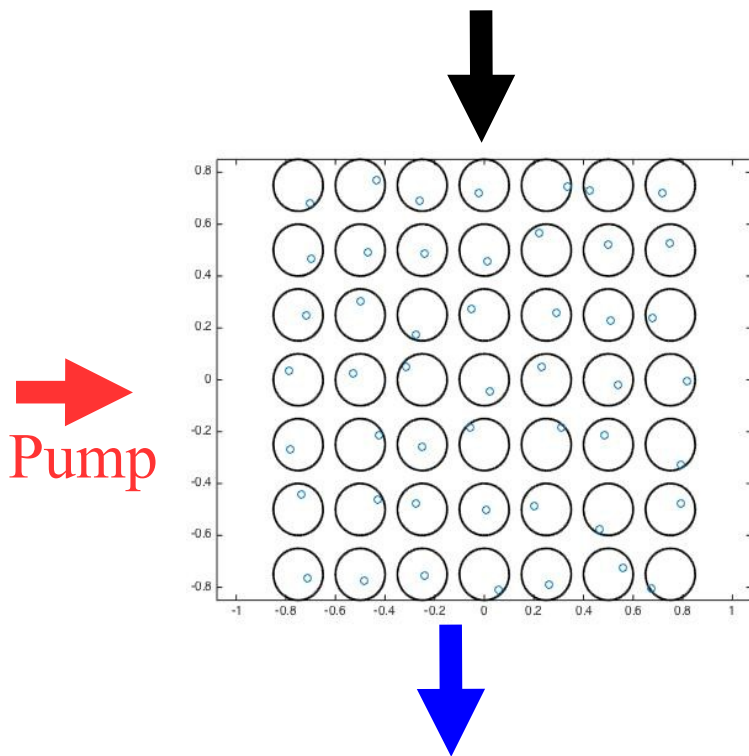
Blue: heterogeneous structure

Red: homogenized structure => effective optical index

No fitting parameters

Low transmission = negative permittivity

Transmission with quantum dots

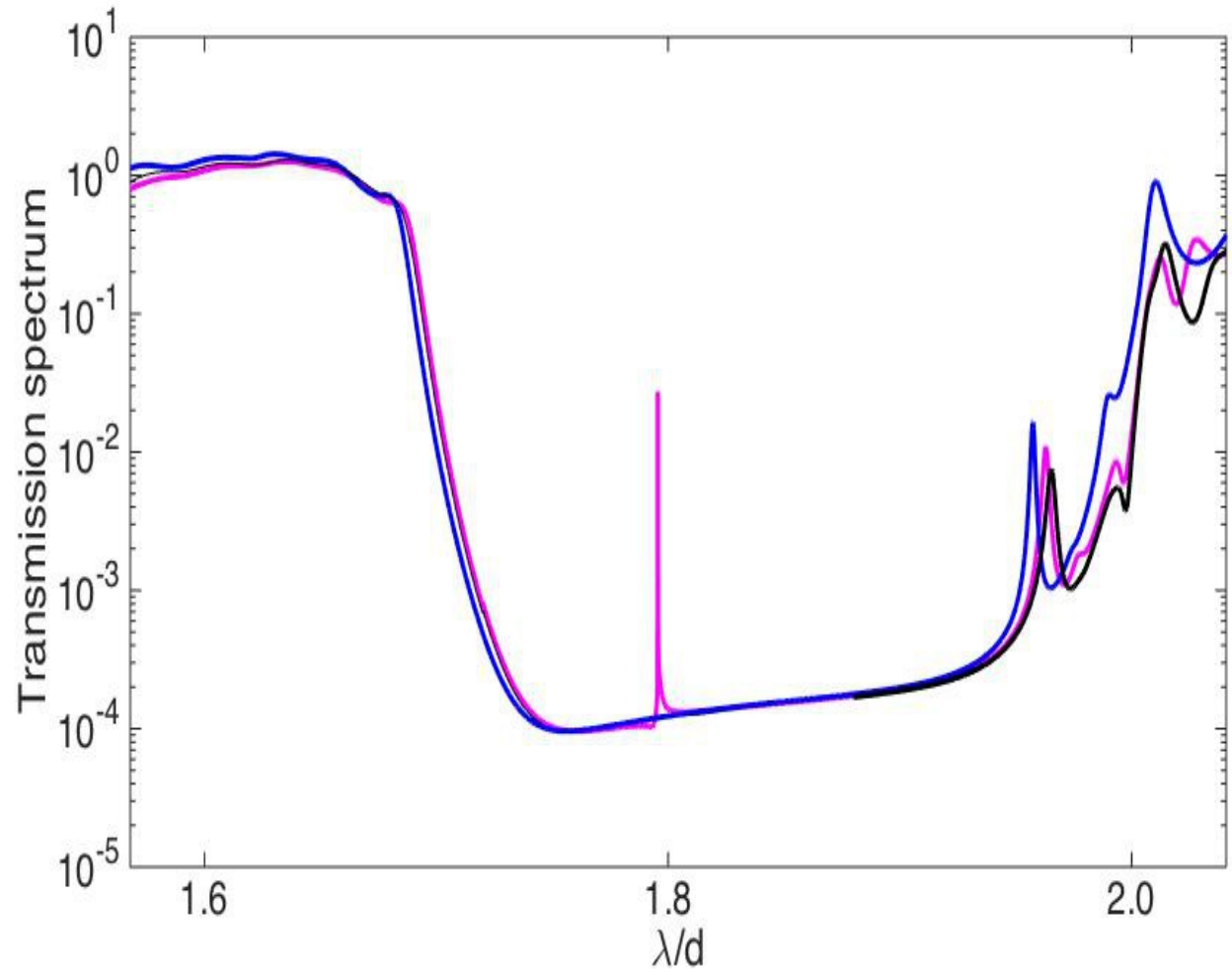


Transmission

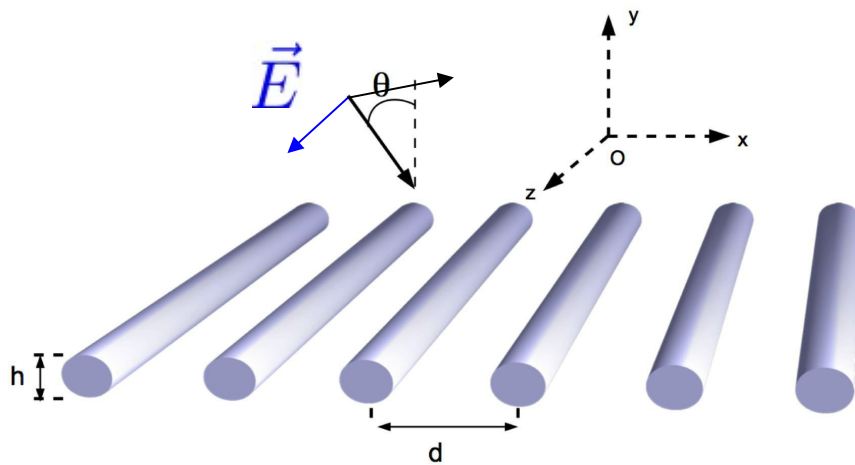
Blue: without pump

Magenta: with pump → quantum dot saturation

Increase of the transmission of light



Single line: metasurface



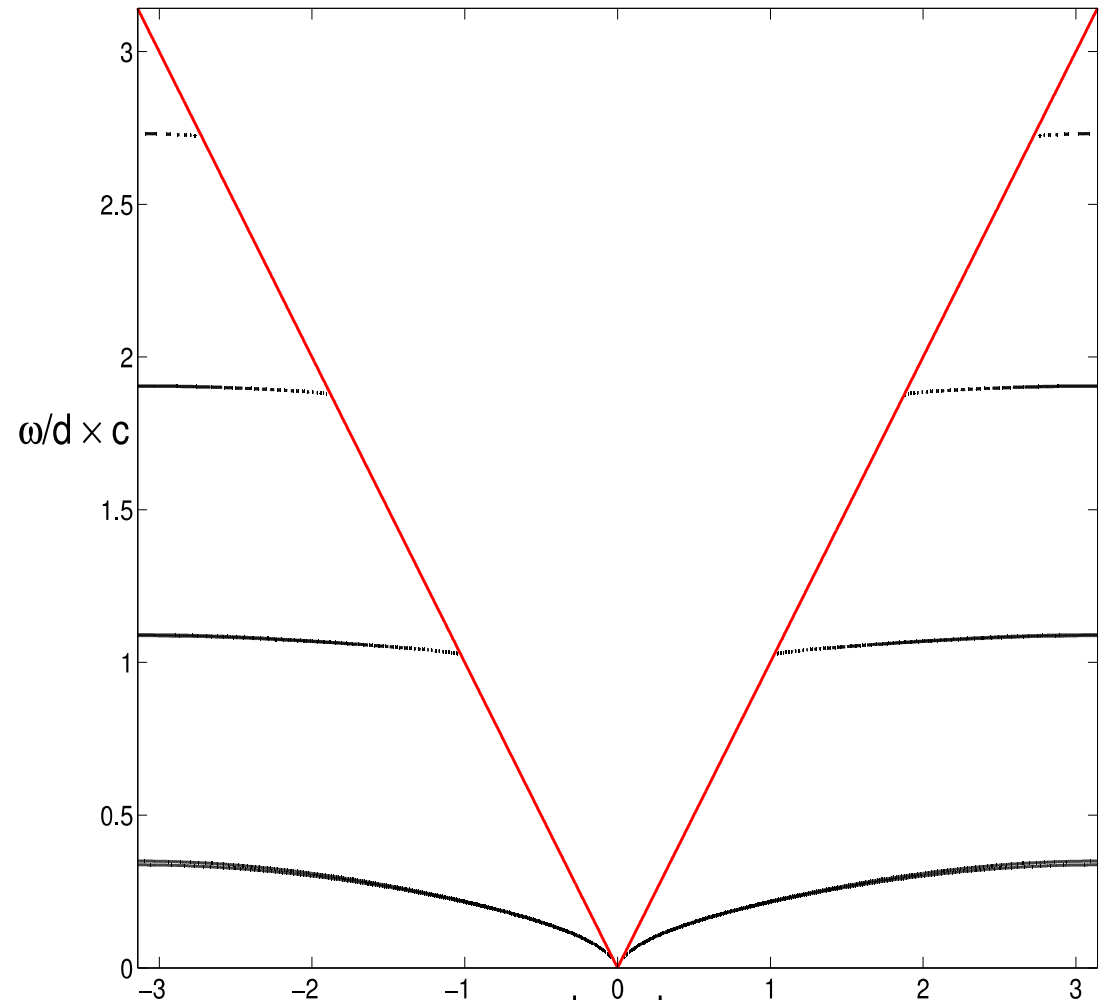
Bloch modes in the structure :

Non-propagating modes :

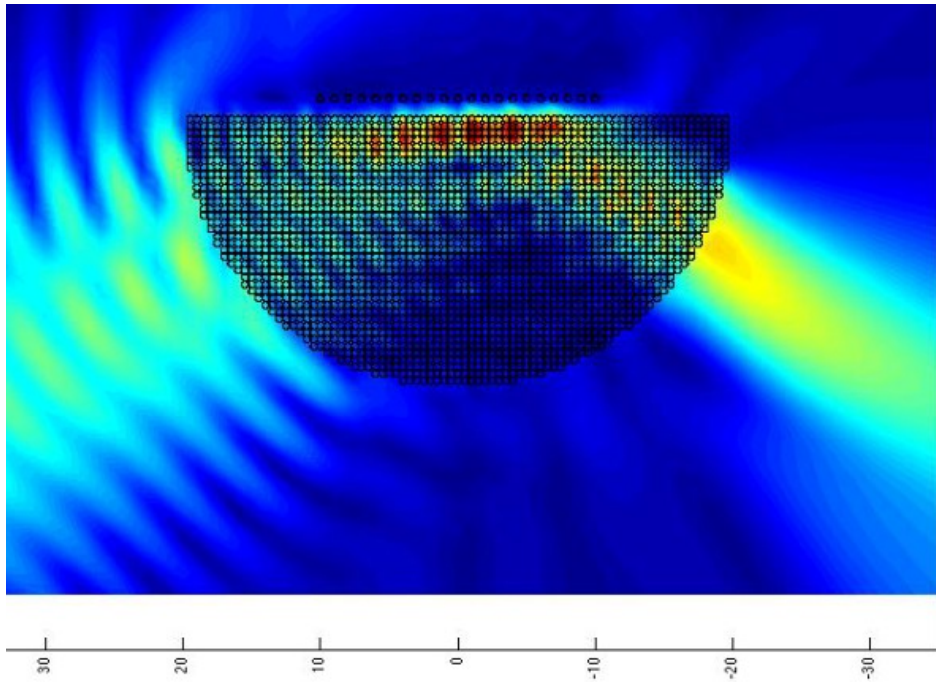
Propagation in x -direction

Evanescent in y-direction

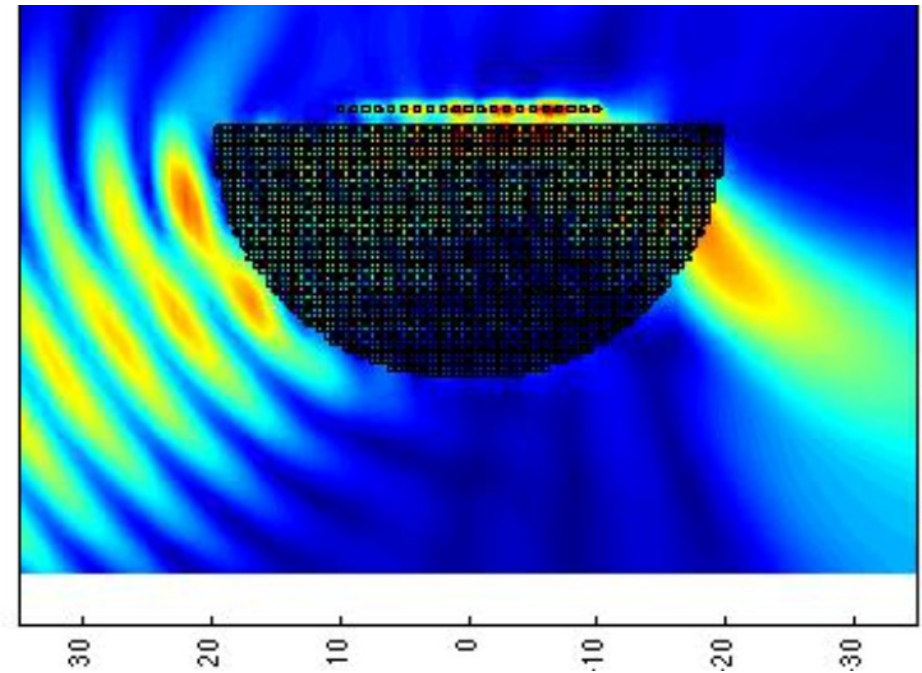
Surface mode in E_{\parallel} polarisation



Exciting the evanescent modes



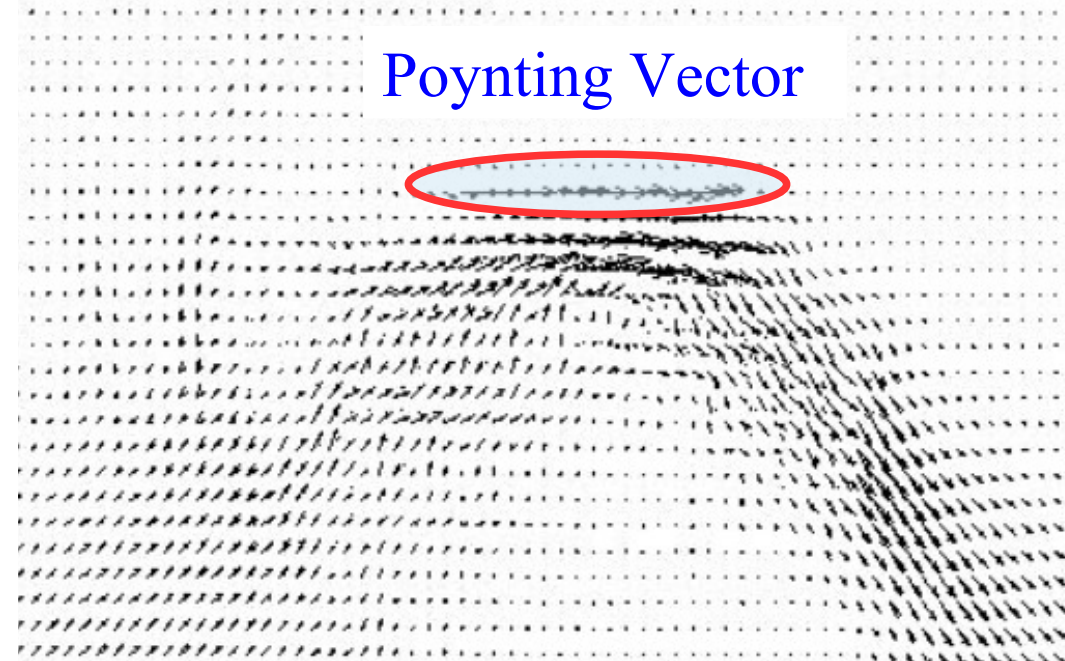
OFF-resonance excitation



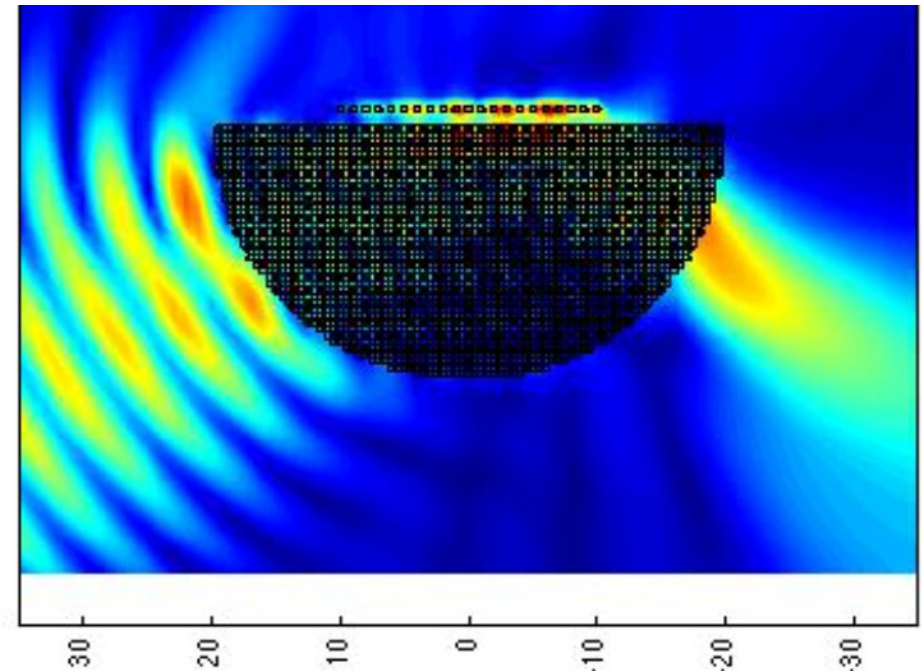
ON-resonance excitation

Exciting the evanescent modes

Poynting Vector



ON-resonance excitation



ON-resonance excitation

Strong coupling between a two-level system and the metasurface
(depend on the distance between atom/metasurface)

Conclusion

Experimental results on a plasmonic meta-surface

Theoretical/Numerical results on a dielectric meta-materials
and a meta-surface without and with quantum-dots

Introduce a quantum description of the quantum dots in the
electromagnetic environment defined by the meta-material and
the meta-surface